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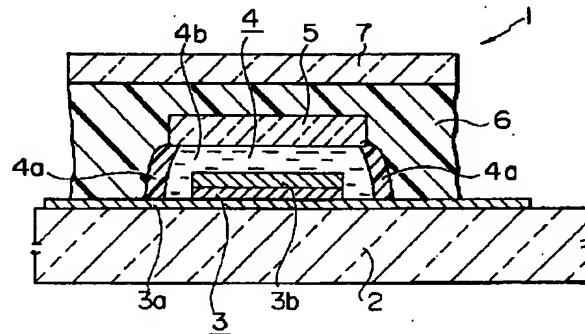
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(54)【発明の名称】 有機ELデバイス

(57)【要約】

【目的】 密着タイプの封止法により封止された有機EL素子を発光源として備えた有機ELデバイスであって、有機EL素子の発光機能が喪失しにくい有機ELデバイスを提供する。

【構成】 本発明の有機ELデバイスは、基板上に設けられた有機EL素子と、この有機EL素子の発光面を覆うようにして前記基板上に形成された接着剤層と、この接着剤層によって前記基板上に固着された封止材とを備えた有機ELデバイスであって、前記有機EL素子と前記接着剤層との間に前記有機EL素子の発光面を覆う応力緩和層を有することを特徴とするものである。



の封止材を接着剤で面接着することにより封止する方法である。

【0007】ケーシングタイプの封止方法によれば、ケース内に入る封止用流体として適当なものを選ぶことにより有機EL素子を破壊することなくその劣化を防止することができるが、その一方で下記(1)～(3)のような難点を生じる。

(1) 有機EL素子をケースに入れることを必須とするので、有機EL素子の利点の1つである薄型という特徴が損なわれる。

(2) ケースを工作する工程およびケース内に封止用流体を注入する工程等が必要であるため、大量生産には向きである。

(3) 基板上に形成されている多数の有機EL素子を1つのケースを用いて封止しようとする場合等のように、大きなケースに大量の封止用液体を入れた場合には、外界の温度上昇に伴う熱膨張によってケース内の封止用流体の体積が増したときにケースを破壊してしまうおそれがある。

【0008】一方、密着タイプの封止方法によれば、封止後においてもその厚さが薄く、かつ大量生産が比較的容易な有機ELデバイス(封止された有機EL素子を発光源として備えているもの)を容易に得ることができる。密着タイプの封止方法としては、GeO等の無機化合物からなる保護膜を封止対象の有機EL素子の外周に設けた後にこの上にガラス基板を固着する方法(特開平4-212284号公報参照)や、封止対象の有機EL素子の外周に直接またはSiO₂膜を介して光硬化性樹脂層を設け、この光硬化性樹脂層を介してガラス基板を固着する方法(特開平5-182759号公報参照)が知られている。

【0009】

【発明が解決しようとする課題】しかしながら、従来の密着タイプの封止方法によって有機EL素子を封止して得た有機ELデバイスには、1000時間程度連続駆動させると有機EL素子の発光機能が失われるという難点があった。

【0010】本発明の第1の目的は、密着タイプの封止法により封止された有機EL素子を発光源として備えた有機ELデバイスであって、有機EL素子の発光機能が喪失しにくい有機ELデバイスを提供することにある。

【0011】また、本発明の第2の目的は、封止後に有機EL素子の発光機能が喪失しにくい有機EL素子の封止方法を提供することにある。

【0012】

【課題を解決するための手段】本発明者らは上述した発光機能の喪失の原因について鋭意検討を加えた結果、この発光機能の喪失は封止効果の低減に起因するものではなく、有機EL素子の接着剤層側の電極端部においてシ

ヨートが発生することに起因していることを見いだした。この点をさらに詳しく述べる。

【0013】接着剤や光硬化性樹脂では硬化時に体積が収縮することから残留応力が発生し、この残留応力は、封止対象の有機EL素子と前記の接着剤や光硬化性樹脂との間にGeOやSiO₂等の膜があったとしてもその膜厚がμmオーダーと薄いと有機ELに伝わってしまう。そして、残留応力は曲率半径の小さな部分で特に強くなるので、有機EL素子に伝播した残留応力は当該素子の端部等に集中する。この結果、素子の電極端部等が押潰され、陽極と陰極とが接触してショートが発生する。本発明者らは上記の知見に基づきさらに検討を加えた結果、本発明に到達した。

【0014】上記第1の目的を達成する本発明の有機ELデバイスは、基板上に設けられた有機EL素子と、この有機EL素子を覆うようにして前記基板上に形成された接着剤層と、この接着剤層によって前記基板上に固着された封止材とを備えた有機ELデバイスであって、前記有機EL素子と前記接着剤層との間に前記有機EL素子を覆う応力緩和層を有することを特徴とするものである。

【0015】また、上記第2の目的を達成する本発明の有機EL素子の封止方法は、基板上に設けられた有機EL素子の背面にこの有機EL素子を覆うようにして接着剤層を設け、この接着剤層によって前記基板上に封止材を固着することにより前記有機EL素子を封止するにあたり、前記有機EL素子と前記接着剤層との間に前記有機EL素子を覆う応力緩和層を設けることを特徴とするものである。

【0016】以下、本発明を詳細に説明する。先ず本発明の有機ELデバイスについて説明すると、この有機ELデバイスは上述したように特定の箇所に応力緩和層を有することを特徴とするものである。そこで、この応力緩和層について先ず説明する。

【0017】上記の応力緩和層は、接着剤層の残留応力が有機EL素子に伝播するのを抑制するためのものである。したがって、この応力緩和層は水分や酸素が外部から有機EL素子に侵入するのを防止する効果(以下、封止効果という)を有していてもよいし有していないてもよいが、封止効果を有して方がより好ましい。また、応力緩和層は必ず応力の発生が小さい物質からなる層のみからなる単層構造の層、または必ず応力の発生が小さい物質からなる層を含む複数層構造の層であることが好ましい。応力緩和層を複数層構造とする場合には、必ず応力の発生が小さい物質からなる層を有機EL素子の外周に形成する。

【0018】ここで、上記の「必ず応力の発生が小さい物質」とは、接着剤層が硬化等の要因により有機EL素子の電極と平行な方向にずれたときに、そのずれを実質的に伝えない性質を有する流体(気体、液体)、グリー

素子の外周のコーティング液が短時間の内に固化してしまうので、前記の補助板を覆うようにして接着剤層を形成する。接着剤層を形成した後にはコーティング液中の溶剤の揮散が抑止されるのでコーティング液の固化が抑止され、その結果として有機EL素子の外周に存在するコーティング液は液体状態を維持し続ける。これにより目的とする応力緩和層が得られる。

【0027】また、前述したグリースを使用しての応力緩和層の形成は、有機EL素子を覆うに十分な量のグリースを有機EL素子上に滴下ないし塗布してグリース層を形成することにより行うことができる。あるいは、前述のようにしてグリース層を形成した後、このグリース層の上に補助板を配置し、この補助板の重みにより、またはこの補助板に荷重をかけることにより前記のグリース層を一様に拡がらせることにより行うことができる。また、補助板を用いずに、グリースを μm オーダーで塗り付けることにより応力緩和層を形成することも可能であるが、 μm オーダーで薄く塗り付けることは、有機EL素子を傷つける可能性もあり、難しいといえる。そして、前述したゲルを使用しての応力緩和層の形成は、グリースを用いて応力緩和層を形成する場合と同様にして行うことができる。

【0028】上述のようにして形成することができる応力緩和層の厚さ（有機EL素子上での厚さ）は、概ね $0.1\mu\text{m} \sim 1\text{cm}$ の範囲内で適宜選択可能である。厚さが $0.1\mu\text{m}$ 未満では所望の効果を得ることが困難である。一方、厚さが 1cm を超えると、薄型という有機ELデバイスの利点が損なわれるばかりでなく、応力緩和層の体積の増大によって熱膨張による体積増も大きくなるので、使用時の温度変化等により破壊するおそれが強くなる。応力緩和層の好ましい厚さは、 $0.01 \sim 1\text{mm}$ である。なお、ここでいう厚さは、応力緩和層が前述したコーティング液を使用して形成したものである場合には有機EL素子の外周に液体状態を維持したまま存在しているコーティング液の層の厚さを意味する。

【0029】また、応力緩和層を形成するにあたって補助板を用いる場合、この補助板は封止効果を有していてもよいし有していないともよいが、封止効果を有している方がより好ましい。補助板としてはガラス、セラミックス、金属等の無機物や、フッ素系樹脂、アクリル系樹脂、ポリカーボネート、ポリエステル、ポリアミド、ポリスチレン、ポリプロピレン、ポリオレフィン系樹脂等の樹脂等でできた板状のものを用いることが好ましい。補助板の大きさは、前述したように平面視したときに有機EL素子を覆うに十分な大きさであることが好ましい。また、補助板の厚さは概ね $0.1\mu\text{m} \sim 3\text{mm}$ とすることが好ましい。厚さが $0.1\mu\text{m}$ 未満のものはその生産コストが高く、また取り扱いにくいことから、望ましくない。一方、厚さが 3mm を超えると薄型という有機ELデバイスの利点が損なわれる。補助板の好ましい

厚さは $0.01 \sim 1\text{mm}$ である。

【0030】本発明の有機ELデバイスでは、基板上に設けられた有機EL素子を覆うようにして上述した応力緩和層が設けられており、この応力緩和層によって覆われた有機EL素子を覆うようにして前記の基板上に接着剤層が形成されている。この接着剤層は後述する封止材を固着させるためのものであると同時に、前述した応力緩和層が外気と触れるのを防止するためのものであり、その側面は特別の部材を設けるか後述する封止材の形状を特別なものとしない限りは外気と接する。したがって、接着剤層は封止効果を有しているもの、すなわち透水性や酸素透過性が低いものであることが特に好ましい。このような接着剤層としては光硬化性接着剤、熱硬化性接着剤、嫌気性接着剤等からなるものが好ましく、具体例としてはベネフィックスVL（（株）アーデル製の光硬化性接着剤の商品名）、アクリルワン#4111（（株）マルト一製の光硬化性接着剤の商品名）、アロンタイトUL（東亜合成化学工業（株）製の嫌気性接着剤の商品名）、アレムコボンド570（（株）オーデック製の熱硬化性接着剤の商品名）等が挙げられる。

【0031】接着剤層は、前述した応力緩和層の上（応力緩和層を形成するにあたって補助板を用いた場合にはこの補助板の上）およびこの応力緩和層を設けた側の基板表面において前記の応力緩和層が設けられていない部分の所望箇所に接着剤を滴下し、この上に後述する封止材を配置し、封止材の重みにより、または封止材に荷重をかけることにより前記の接着剤を一様に拡がらせた後に当該接着剤を硬化させることにより形成することができる。また、塗布法、スピントコート法、ディップ法等の方法により未硬化の接着剤層を形成した後、この接着剤層の上に後述する封止材を配置してから当該接着剤を硬化させることによっても形成することができる。

【0032】接着剤層の最終的な厚さ（応力緩和層上の厚さ。応力緩和層を形成するにあたって補助板を用いた場合にはこの補助板上での厚さ）は概ね $0.1\mu\text{m} \sim 1\text{cm}$ とすることが好ましい。厚さが $0.1\mu\text{m}$ 未満では、封止材を載せたときに当該封止材が未硬化状態の接着剤の表面張力により浮き上がろうとする結果、未硬化状態の接着剤の中やこの接着剤と封止材との界面に気泡が入り易い。一方、厚さが 1cm を超えると、薄型という有機ELデバイスの利点が損なわれる。接着剤層の好ましい厚さは $0.001 \sim 1\text{mm}$ である。接着剤層の形成時においては、有機EL素子が応力緩和層によって被覆されていることから、接着剤の硬化時の硬度や収縮率に注意を払う必要が実質的がない。その結果として、応力緩和層が無い場合よりも接着剤の選択の幅が広がる。

【0033】上述した接着剤層によって基板（有機EL素子が設けられているもの）上に固着される封止材は、外部から有機EL素子に水分や酸素が侵入を防止するためのものであるので、封止効果の高いもの、すなわち透

ロン誘導体、フェニレンジアミン誘導体、アリールアミン誘導体、アミノ置換カルコン誘導体、オキサゾール誘導体、スチリルアントラセン誘導体、フルオレノン誘導体、ヒドラゾン誘導体、スチルベン誘導体、シラザン誘導体、ポリシラン系化合物、アニリン系共重合体、チオフェンオリゴマー等の導電性高分子オリゴマー、ポルフィリン化合物、芳香族第三級アミン化合物、スチリルアミン化合物、芳香族ジメチリディン系化合物等が挙げられる。正孔注入層の厚さも特に限定されるものではないが、通常は5nm～5μmの範囲内で適宜選択される。正孔注入層は上述した材料の1種または2種以上からなる一層構造であってもよいし、同一組成または異種組成の複数層からなる複数層構造であってもよい。

【0043】電子注入層は陰極から注入された電子を発光層に伝達する機能を有していればよく、その材料（電子注入材料）の具体例としては、ニトロ置換フルオレノン誘導体、アントラキノジメタン誘導体、ジフェニルキノン誘導体、チオビランジオキシド誘導体、ナフタレンペリレン等の複素環テトラカルボン酸無水物、カルボジイミド、フレオレニリデンメタン誘導体、アントラキノジメタン誘導体、アントロン誘導体、オキサジアゾール誘導体、8-キノリノール誘導体の金属錯体、メタルフリーフタロシアニンやメタルフタロシアニンあるいはこれらの末端がアルキル基やスルホン基等で置換されているもの、ジスチリルピラジン誘導体等が挙げられる。電子注入層の厚さも特に限定されるものではないが、通常は5nm～5μmの範囲内で適宜選択される。電子注入層は上述した材料の1種または2種以上からなる一層構造であってもよいし、同一組成または異種組成の複数層からなる複数層構造であってもよい。

【0044】そして、保護層の材料の具体例としては、テトラフルオロエチレンと少なくとも1種のコモノマーとを含むモノマー混合物を共重合させて得られる共重合体、共重合主鎖に環状構造を有する含フッ素共重合体、ポリエチレン、ポリプロピレン、ポリメチルメタクリート、ポリイミド、ポリユリア、ポリテトラフルオロエチレン、ポリクロロトリフルオロエチレン、ポリジクロロジフルオロエチレン、クロロトリフルオロエチレンとジクロロジフルオロエチレンとの共重合体、吸水率1%以上の吸水性物質および吸水率0.1%以下の防湿性物質、In, Sn, Pb, Au, Cu, Ag, Al, Ti, Ni等の金属、MgO, SiO, SiO₂, Al₂O₃, GeO, NiO, CaO, BaO, Fe₂O₃, Y₂O₃, TiO₂等の金属酸化物、MgF₂, LiF, AlF₃, CaF₂等の金属フッ化物等が挙げられる。

【0045】また、有機EL素子を構成する各層（陽極および陰極を含む）の形成方法についても特に限定されるものではない。陽極、陰極、発光層、正孔注入層、電子注入層の形成方法としては、例えば真空蒸着法、スピ

シコート法、キャスト法、スパッタリング法、LB法等を適用することができるが、発光層についてはスパッタリング法以外の方法（真空蒸着法、スピコート法、キャスト法、LB法等）を適用することが好ましい。発光層は、特に分子堆積膜であることが好ましい。ここで分子堆積膜とは、気相状態の材料化合物から沈着され形成された薄膜や、溶液状態または液相状態の材料化合物から固化され形成された膜のことであり、通常この分子堆積膜は、LB法により形成された薄膜（分子累積膜）とは凝集構造、高次構造の相違や、それに起因する機能的な相違により区分することができる。スピコート法等により発光層を形成する場合には、樹脂等の接着剤と材料化合物とを溶剤に溶かすことによりコーティング溶液を調製する。

【0046】また、保護層については真空蒸着法、スピシコート法、スパッタリング法、キャスト法、MBE（分子線エピタキシ）法、クラスターイオンビーム法、イオンプレーティング法、プラズマ重合法（高周波励起イオンプレーティング法）、反応性スパッタリング法、プラズマCVD法、レーザーCVD法、熱CVD法、ガスソースCVD法等を適用することができる。

【0047】各層の形成方法は、使用的する材料に応じて適宜変更可能である。有機EL素子を構成する各層の形成にあたって真空蒸着法を用いれば、この真空蒸着法だけによって有機EL素子を形成することができるため、設備の簡略化や生産時間の短縮を図るうえで有利である。

【0048】なお、有機EL素子を構成する各層（保護層を含む）の形状および大きさは必ずしも同一ではない。また、素子を平面視したときに、基板の直上に形成された電極の上に他の全ての層が必ず納まっているというわけでもない。一方、本発明でいう応力緩和層は接着剤層の残留応力が有機EL素子に伝播して当該有機EL素子にショートが生じるのを抑制するためのものである。したがって、本発明でいう「有機EL素子を覆う応力緩和層」とは、有機EL素子を平面視したときに陰極と陽極とが重なる領域を少なくとも覆っている応力緩和層を意味する。また、本明細書でいう「有機EL素子を覆うに十分な量のコーティング液（グリース、ゲル）」とは、「有機EL素子を平面視したときに陰極と陽極とが重なる領域を少なくとも覆うに十分な量のコーティング液（グリース、ゲル）」を意味する。そして、「平面視したときに有機EL素子を覆うに十分な大きさを有する補助板」とは「平面視したときに、有機EL素子を平面視したときに陰極と陽極とが重なる領域を少なくとも覆うに十分な大きさを有する補助板」を意味する。

【0049】また、本発明の有機ELデバイスを構成する有機EL素子の数は1個であってもよいし複数個であってもよい。複数個とする場合、各素子は層構成や発光色から見た種類が同一であってもよいし異なっていても

ニウム（以下、Al_{q3}という）を200mg入れて、真空チャンバー内を 1×10^{-4} Paまで減圧した。

【0058】次に、TPDを入れた前記の抵抗加熱ポートを215~220℃まで加熱し、TPDを蒸着速度0.1~0.3nm/秒でITO膜上に堆積させて、膜厚60nmの正孔注入層を成膜した。このときの基板温度は室温であった。次いで、正孔注入層が成膜された透明支持基板を真空チャンバーから取出すことなく、正孔注入層の成膜に引続いて発光層の成膜を行った。発光層の成膜は、Al_{q3}を入れた前記の抵抗加熱ポートを275℃まで加熱し、Al_{q3}を蒸着速度0.1~0.2nm/秒で正孔注入層上に堆積させて、膜厚60nmのAl_{q3}層を成膜することにより行った。このときの基板温度も室温であった。次に、モリブデン製抵抗加熱ポートにマグネシウム1gを入れ、別のモリブデン製抵抗加熱ポートに銀500mgを入れて、真空チャンバー内を 2×10^{-4} Paまで減圧した。そして、マグネシウムを入れた前記の抵抗加熱ポートを500℃程度に加熱してマグネシウムを約1.7~2.8nm/秒の蒸着速度で蒸発させると共に、銀を入れた前記の抵抗加熱ポートを800℃程度に加熱して銀を0.03~0.08nm/秒の蒸着速度で蒸発させて、マグネシウムと銀との混合金属からなる膜厚150nmの陰極を発光層上に計3個設けた。各陰極は平面視上の大きさが3mm×15mmのものであり、これらは一定間隔でITO膜と直交する向きに設けられている。

【0059】この後、前記の陰極まで設けたガラス基板を3つに切り、ガラス基板上の層構成が陽極（ITO膜）/正孔注入層/発光層/陰極（Mg・Ag層）である有機EL素子を計3つ得た。これらの有機EL素子の発光面（平面視したときに陰極と陽極が重なり、これらの電極の間に正孔注入層および発光層が存在している領域）の大きさは3mm×5mmである。また、これらの有機EL素子の初期輝度は電圧6.5V、電流密度3mA/cm²の条件下で100cd/m²に達し、このときの電力変換効率は1.6lm/Wであった。

【0060】(2) 応力緩和層および接着剤層の形成並びに封止材の配設

先ず、応力緩和層の材料としてフッ素樹脂コーティング液（旭硝子（株）製のサイトップCTX-105A）を用意し、また、補助板として約10mm×10mm×0.15mmの大きさのカバーガラス（以下、カバーガラスIという）を用意した。次に、前記のフッ素樹脂コーティング液5μlを上記(1)で作製した有機EL素子上に滴下し、この上に前記のカバーガラスIを載せた。フッ素樹脂コーティング液の粘性の低さとカバーガラスIの重みにより前記コーティング液は一様に広がり、有機EL素子とカバーガラスIとの間には前記コーティング液の薄い膜（厚さ0.05mm）ができた。このとき、コーティング液の一部はカバーガラスIによっ

て覆われる領域の外側に若干はみ出した。この状態で10分程放置することにより、フッ素樹脂コーティング液からなる上記の薄い膜の外表面、すなわちガラス基板の表面と平行な方向の表面に固体層（被膜）を形成させて、目的とする応力緩和層を一応得た。このとき、有機EL素子に接している部分のフッ素樹脂コーティング液は固化せずに液体状態を保っていた。

【0061】次に、接着剤として光硬化性接着剤

(（株）アーデル製のベネフィックスVL) 0.1mlを上記のカバーガラスI上に滴下し、その上に封止材としてのカバーガラス（大きさは約20mm×20mm×0.15mm；以下、カバーガラスIIという）を載せた。光硬化性接着剤の粘性の低さとカバーガラスIIの重みにより前記光硬化性接着剤は一様に広がり、上記の応力緩和層およびカバーガラスIを完全に覆った。カバーガラスIIの載置に引き続き、ハロゲンランプの光を前記のカバーガラスII上から光硬化性接着剤に照射して、当該光硬化性接着剤を硬化させた。

【0062】これにより、上記の応力緩和層を覆うようにして光硬化性接着剤からなる厚さ0.1mm（カバーガラスI上での厚さ）の接着剤層が形成され、同時に前記の接着剤層によって封止材としてのカバーガラスIIがガラス基板（有機EL素子が設けられているもの）上に固着された。さらに、接着剤層を形成した後には上記の応力緩和層中のフッ素樹脂コーティング液の溶剤の揮散が抑制されるので、有機EL素子の外周に存在するフッ素樹脂コーティング液は液体状態を維持し続け、その結果として目的とする応力緩和層が最終的に形成された。

【0063】上述のようにして接着剤層の形成および封止材の配設まで行ったことにより、目的とする有機ELデバイスが得られた。以下、同様にして有機ELデバイスを作製し、計45個の有機ELデバイスを得た。本実施例1で作製した有機ELデバイスの断面の概略図を図1に、また上面図を図2に示す。

【0064】図1に示したように、この有機ELデバイス1はガラス基板2の片面に形成された有機EL素子3と、この有機EL素子3を覆うようにしてガラス基板2上に形成された応力緩和層4と、この応力緩和層4を形成するために用いたカバーガラスIからなる補助板5と、この補助板5および前記応力緩和層4を覆うようにしてガラス基板2上に形成された光硬化性接着剤からなる接着剤層6と、この接着剤層6によってガラス基板2上に固着されたカバーガラスIIからなる封止材7とを備えたものである。応力緩和層4はフッ素樹脂コーティング液を材料とするものであり、この応力緩和層4においてガラス基板2の表面と平行な方向の表面はその形成時に溶剤が蒸発したことにより固化して固体層4aを形成しており、有機EL素子3に接している部分のフッ素樹脂コーティング液4bは固化せずに液体状態を保っている。

(以下、注入口という)とを有するガラス板(外寸: 20 mm × 75 mm × 3 mm、以下シールドガラスという)を用意し、このシールドガラスと保護層まで成膜した有機EL素子とをエポキシ系接着剤(セメダイン社製のセメダインスーパー-5)により貼り合わせた。エポキシ系接着剤は、主剤と硬化剤とを混ぜ合わせてヘラで20回かき混ぜてから、上記保護層まで成膜した有機EL素子の縁部に幅1 mmではほぼ20 × 75 mmの長方形に塗布した。またシールドガラスと上記保護層まで成膜した有機EL素子とは、陰極および保護層がシールドガラスの凹部内に収まるようにして貼り合わせた。貼り合わせ後、大気中に10時間放置してエポキシ系接着剤を硬化させた。

【0075】次いで、シールドガラスに設けられている注入口から、吸湿用のシリカゲル(粒径50 μm)を8体積%分散させたシリコーンオイル(株)東芝製のTSK451、以下封止用液体という)を注入して、シールドガラスの凹部と上記保護層まで成膜した有機EL素子とにより形成された空間内を封止用液体で満たした。

この後、注入口をガラス製の蓋で封鎖して、ケーシングタイプの封止が施された有機ELデバイスを得た。なお、ガラス製の蓋は、上述したエポキシ系接着剤によりシールドガラスに接着させた。以下、同様にして有機ELデバイスを作製し、計45個の有機ELデバイスを得た。

【0076】歩留りの評価

実施例1～実施例3および比較例1～比較例3でそれぞれ作製した有機ELデバイスについて、デバイス作製直後から初期輝度が100 cd/m²となる直流定電流で有機EL素子を連続駆動し、連続駆動開始直後、1週間後、1箇月(31日間)後および1000時間後のそれぞれの時点で、ショートにより発光機能を喪失した有機EL素子の数を数えた。なお、輝度の測定は輝度計(ミノルタ社製のCS-100)を用いて大気中で行った。この結果を表1に示す。

【0077】

【表1】

表 1

	評価に付した デバイスの数	発光機能を喪失した素子の数			
		駆動開始直後	1週間後	1箇月後	1000時間後
実施例1	45	1	1	1	1
実施例2	45	3	3	3	3
実施例3	45	4	4	4	4
比較例1	45	43	45	45	45
比較例2	45	2	8	31	45
比較例3	45	1	2	2	3

【0078】表1に示したように、実施例1～実施例3でそれぞれ作製した有機ELデバイスではショートにより発光機能を喪失した有機EL素子の数が少ない。このことから、信頼性の高い有機ELデバイスが高い歩留まりの下に作製されたことがわかる。これに対し、応力緩和層を設けなかった比較例1では連続駆動開始直後の時点での有機EL素子がショートにより発光機能を喪失しており、1週間後には全ての有機EL素子がショートにより発光機能を喪失していた。このことから、応力緩和層は有機EL素子がショートによってその発光機能を喪失するのを防止するうえで有用であることがわかる。

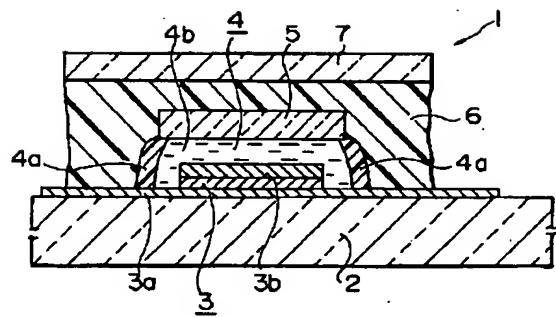
【0079】また、比較例2で作製した有機ELデバイスでは、連続駆動開始直後の時点ではショートによって発光機能を喪失した有機EL素子の数が少ないが、1000時間後には全ての素子がショートにより発光機能を

喪失していた。そして、比較例3で作製した有機ELデバイスでは、全ての調査時点で、ショートによって発光機能を喪失した有機EL素子の数が実施例1～実施例3と同等かそれ以下である。したがって、比較例3では有機EL素子がショートによってその発光機能を喪失するか否かという観点からみた信頼性が高い有機ELデバイスが高い歩留まりの下に作製されたことができ、この点で比較例3の有機ELデバイスは優れたものである。しかしながら、後述する封止効果や製造の難易、有機ELデバイスの厚さ等をも考慮すると、実施例1～実施例3の有機ELデバイスの方が優れているといえる。

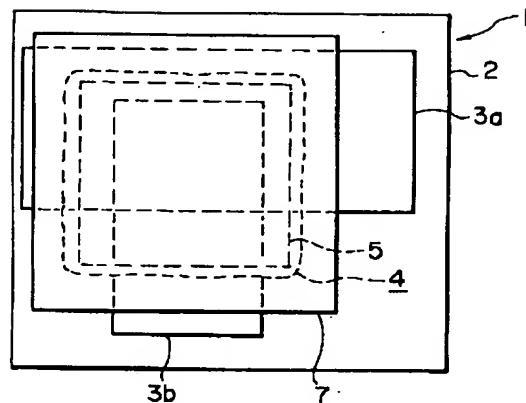
【0080】封止効果の評価

有機EL素子に見られるダーケスポットと呼ばれる丸い形の無発光領域は、有機EL素子に水分や酸素が侵入することにより拡大していく。したがって、任意のダークスポットについてその大きさの経時変化を調べること

【図1】



【図2】



PATENT ABSTRACTS OF JAPAN

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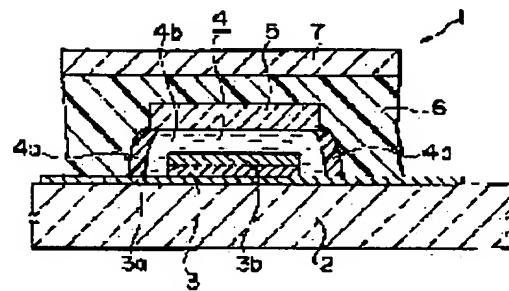
(72)Inventor : FUKUOKA KENICHI

FUJITA MASATO

(54) ORGANIC EL DEVICE**(57)Abstract:**

PURPOSE: To provide a thin organic EL device stable over a long period in a high yield with a relatively easy manufacturing process by providing a stress moderating layer for covering the emitting surface of an organic EL element between an adhesive layer and the organic EL element.

CONSTITUTION: An adhesive layer 6, for example, 0.1mm in thickness consisting of an optical hardening adhesive is formed so as to cover a stress relaxation layer 4, and a cover glass 1 as a sealing material is fixed onto a glass substrate 2 by the adhesive layer 6. Further, since the evaporation of a solvent of fluorine resin coating solution in the stress moderating layer 4 is suppressed after the adhesive layer 6 is formed, the fluorine resin coating solution present on the outer circumference of an organic EL element 3 continues to keep the liquid state, and as the result, the intended stress relaxation layer 4 is finally formed. The stress relaxation layer 4 for covering the adhesive layer 6 and the emitting surface of the organic EL element 3 is provided, whereby a thin organic EL device 1 stable over a long period can be provided with a relatively easy manufacturing process in a high yield.

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CLAIMS**[Claim(s)]**

[Claim 1] The organic electroluminescence device characterized by having a wrap stress relaxation layer for said organic EL device between said organic EL devices and said adhesives layers in the organic electroluminescence device equipped with the sealing agent which fixed on said substrate by the organic EL device formed on the substrate, the adhesives layer formed on said substrate as covered this organic EL device, and this adhesives layer.

[Claim 2] The organic electroluminescence device according to claim 1 with which this layer is formed in the periphery of an organic EL device including the layer which generating of shear stress becomes from the matter with a small stress relaxation layer.

[Claim 3] The organic electroluminescence device according to claim 2 with which the matter with small generating of shear stress consists of the liquid of a fluorine system, a silicone system, or a hydrocarbon system, grease, or gel.

[Claim 4] An organic electroluminescence device given in any 1 term of claim 1 – claim 3 which a stress relaxation layer becomes from the solidification object of the liquid of the same presentation as the liquid with which it has the liquid layer formed in the periphery of an organic EL device, and the solid-state layer formed in the periphery of this liquid layer, and said solid-state layer forms said liquid layer.

[Claim 5] An organic electroluminescence device given in any 1 term of claim 1 – claim 4 prepared in the sense to which an organic EL device is countered as substantially [a wrap accessory plate] as a substrate when plane view is carried out from a tooth back between a stress relaxation layer and an adhesives layer.

[Claim 6] The organic electroluminescence device according to claim 5 with which a stress relaxation layer consists of a solidification object of the liquid of the same presentation as the liquid with which it has the liquid layer formed in the periphery of the luminescence side of an organic EL device, and the solid-state layer formed in the periphery of a direction parallel to a substrate front face in this liquid layer, and said solid-state layer forms said liquid layer.

[Claim 7] The closure approach of the organic EL device characterized by preparing an adhesives layer in it as this organic EL device is covered at the tooth back of an organic EL device prepared on the substrate, and forming said organic EL device for a wrap stress relaxation layer between said organic EL devices and said adhesives layers in closing said organic EL device by fixing a sealing agent on said substrate by this adhesives layer.

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the closure approach of the organic electroluminescence device equipped with the organic EL device as a source of luminescence, and an organic EL device, and relates to the adhesion type closure approach for closing the organic electroluminescence device and organic EL device which were equipped with the organic electroluminescence by which the closure was especially carried out by the adhesion type closure approach as a source of luminescence.

[0002]

[Description of the Prior Art] There are an inorganic EL element and an organic EL device in an EL element, since any EL element is self-luminous, visibility is high, and handling is easy while excelling in shock resistance, since it is a perfect solid-state component. For this reason, the researches and developments and utilization as the pixel of graphic display, the pixel of a television image display device, or the surface light source are advanced. An organic EL device makes the luminous layer which contains an organic luminescent material for an anode plate and cathode among these intervene at least, and carries out a laminating, and the thing of various lamination is developed by current. As an example of lamination, an anode plate / luminous layer / cathode, an anode plate / luminous layer / electron injection layer / cathode, an anode plate / hole-injection layer / luminous layer / cathode, an anode plate / hole-injection layer / luminous layer / electron injection layer / cathode, etc. are mentioned. Although a luminous layer is usually formed of one sort or two or more sorts of organic luminescent material, it may be formed with mixture with an organic luminescent material, a hole-injection ingredient, and/or an electron injection ingredient.

[0003] In many cases, the organic EL device which has such lamination is formed on a substrate. And when taking out EL light from a luminous layer from the aforementioned substrate side, an anode plate is formed in right above [of a substrate], and cathode is formed in right above [of a substrate] when taking out EL light from a luminous layer from the side which formed the organic EL device. An anode plate is formed with the transparency thru/or the translucent thin film which consists of big matter of work functions, such as Au, nickel, and ITO (indium stannic acid ghost). On the other hand, cathode is formed with the thin film which consists of small matter of work functions, such as Yb, Mg, aluminum, and In.

[0004] An organic EL device is a light emitting device of a current drive mold, and uses luminescence produced when the electron and electron hole which were poured into the luminous layer (organic luminescent material) recombine. For this reason, the organic EL device has the advantage that a drive by low battery called 4.5V is possible by making thickness of a luminous layer thin, and a response is also quick, and the advantage that luminescence of high brightness is possible since brightness is proportional to an inrush current. moreover, luminescence of various colors, such as 4 gloss blue, green, yellow, and red, is obtained by changing luminescent material. The research for current and utilization is continued by these advantages.

[0005] By the way, the organic substance, such as an organic luminescent material used for an organic EL device, is weak into moisture, oxygen, etc. Moreover, also in an electrode, in atmospheric air, a property deteriorates rapidly by oxidation. Therefore, in order to obtain a practical organic EL device and an organic electroluminescence device, it is necessary to give the closure. As the closure approach of an organic EL device, it is SiO₂ to the approach (refer to JP,4-137483,A) of forming the poly paraxylene film as a closure layer in the component front face for the closure by the vapor-phase-polymerization method, and the component front face for the closure. Although there is also the approach (refer to JP,4-73886,A) of forming a protective coat, the approach that the closure effectiveness is more high is already developed. The approach is divided roughly into the following two gestalten.

[0006] One is the casing type closure approach and it is the approach of closing by putting in an organic EL device in a case, intercepting with the external world, and being filled up with the predetermined fluid for the closures with the organic EL device in the aforementioned case. Another is the adhesion type closure approach and is the approach of closing by carrying out face bonding of the sealing agents, such as a glass plate, to the tooth back (in view of a substrate side back of a component) of the organic EL device currently formed on the substrate with adhesives.

[0007] According to the casing type closure approach, the degradation can be prevented, without destroying an organic EL device by choosing a thing suitable as a fluid for the closures put in in a case, but on the other hand, a difficulty like following the (1) - (3) is produced.

- (1) Since it makes it indispensable to put an organic EL device into a case, the description of the thin shape which is one of the advantages of an organic EL device is spoiled.

(2) Since the process which pours in fluid for the closures is required in the process which machines a case, and a case, it is unsuitable for mass production method.

(3) Like [when it is going to close the organic EL device of a large number currently formed on the substrate using one case], when a lot of liquids for the closures are put into a big case, and the volume of the fluid for the closures within a case increases by the thermal expansion accompanying the temperature rise of the external world, there is a possibility of destroying a case.

[0008] On the other hand, according to the adhesion type closure approach, an organic electroluminescence device (what is equipped with the organic EL device by which the closure was carried out as a source of luminescence) with comparatively easy mass production method can be easily obtained thinly [the thickness] after the closure, and the high closure effectiveness can be acquired easily. They are direct or SiO₂ to the periphery of the approach (refer to JP,4-212284,A) of fixing a glass substrate on this, after preparing the protective coat which consists of inorganic compounds, such as GeO, in the periphery of the organic EL device for the closure as the adhesion type closure approach, and the organic EL device for the closure. A photo-setting resin layer is prepared through the film, and the approach (refer to JP,5-182759,A) of fixing a glass substrate through this photo-setting resin layer is learned.

[0009]

[Problem(s) to be Solved by the Invention] However, when the organic electroluminescence device which closed and obtained the organic EL device by the closure approach of adhesion type [conventional] was made to carry out a continuation drive for about 1000 hours, there was a difficulty that the luminescence function of an organic EL device is lost in it.

[0010] The 1st purpose of this invention is the organic electroluminescence device equipped with the organic EL device by which the closure was carried out by the adhesion type closure method as a source of luminescence, and is to offer the organic electroluminescence device which the luminescence function of an organic EL device cannot lose easily.

[0011] Moreover, the 2nd purpose of this invention is to offer the closure approach of the organic EL device which the luminescence function of an organic EL device cannot lose easily after the closure.

[0012]

[Means for Solving the Problem] As a result of this invention persons' adding examination wholeheartedly about the cause of loss of the luminescence function mentioned above, it found out loss of this luminescence function not originating in reduction of the closure effectiveness, and originating in short-circuit occurring in the electrode edge by the side of the adhesives layer of an organic EL device. This point is explained in more detail.

[0013] between the adhesives of the organic EL device and the above for the closure, or photo-setting resins — GeO and SiO₂ etc. — even if there is film, if that thickness is thin, it will get across to organic electroluminescence with mum order. [since the volume contracts in adhesives or a photo-setting resin at the time of hardening residual stress occurs, and] [this residual stress] And since residual stress becomes strong especially in a part with small radius of curvature, the residual stress spread to the organic EL device is concentrated on the edge of the component concerned etc. Consequently, the crush of the electrode edge of a component etc. is carried out, an anode plate and cathode contact, and short-circuit occurs. this invention persons reached this invention, as a result of adding examination further based on the above-mentioned knowledge.

[0014] The organic electroluminescence device of this invention which attains the 1st purpose of the above is an organic electroluminescence device equipped with the organic EL device formed on the substrate, the adhesives layer formed on said substrate as covered this organic EL device, and the sealing agent which fixed on said substrate by this adhesives layer, and is characterized by having a wrap stress relaxation layer for said organic EL device between said organic EL devices and said adhesives layers.

[0015] Moreover, the closure approach of the organic EL device of this invention of attaining the 2nd purpose of the above is characterized by preparing an adhesives layer in it, as this organic EL device is covered at the tooth back of an organic EL device prepared on the substrate, and forming said organic EL device for a wrap stress relaxation layer between said organic EL devices and said adhesives layers in closing said organic EL device by fixing a sealing agent on said substrate by this adhesives layer.

[0016] Hereafter, this invention is explained to a detail. If the organic electroluminescence device of this invention is explained first, this organic electroluminescence device will be characterized by having a stress relaxation layer in a specific part, as mentioned above. Then, this stress relaxation layer is explained first.

[0017] The above-mentioned stress relaxation layer is for controlling that the residual stress of an adhesives layer spreads to an organic EL device. Therefore, although this stress relaxation layer may have the effectiveness (henceforth the closure effectiveness) of preventing moisture and oxygen invading into an organic EL device from the exterior and does not need to have it, it has the closure effectiveness and the direction is more desirable. Moreover, as for a stress relaxation layer, it is desirable that they are the layer of the monolayer structure which consists only of a layer which generating of shear stress becomes from the small matter, and the layer of two or more layer structure containing the layer which generating of blank stress becomes from the small matter. In making a stress relaxation layer into two or more layer structure, it forms in the periphery of an organic EL device the layer which generating of shear stress becomes from the small matter.

[0018] Here, the above-mentioned "matter with small generating of shear stress" means the fluid (a gas, liquid) which has the property in which the gap is not told substantially, grease, gel, etc., when an adhesives layer shifts in the direction parallel to the electrode of an organic EL device according to factors, such as hardening.

[0019] Generating of the above-mentioned shear stress can mention hydrocarbon system liquids, such as silicone system liquids, such as fluorine system liquids, such as gases, such as gaseous helium, nitrogen gas, and argon gas, and chlorofluorocarbon, a perfluoro polyether, a perfluoro amine, a perfluoro alkane, methyl hydrogen silicone, methyl chlorophenyl silicone, and trifluoro propylmethyl silicone, an alpha olefin (Pori alpha olefin of C5 – C17), polybutene, alkylbenzene, and a polyalkylene glycol, etc. as a fluid among small matter. However, a complicated process is needed for forming, preventing that the adhesives for adhesives stratification advance the stress relaxation layer which consists of these fluids into the inside of the stress relaxation layer concerned, or the interface of the stress relaxation layer and a substrate (substrate for forming an organic EL device) concerned. Therefore, when using a fluid as the above-mentioned "matter with small generating of shear stress", it is desirable to use the coating liquid which is made to come to dissolve resin, rubber, etc. in a solvent as this fluid, and carries out film formation by evaporation of a solvent under about 10–30-degree C temperature conditions among atmospheric air.

[0020] When the above-mentioned coating liquid is used, it is possible to be able to make a solid-state layer (coat) form in a layer front face, and to maintain a liquid condition about the part inside the aforementioned solid-state layer by evaporating a little the solvent in the layer which consists of the coating liquid concerned. After forming the layer which consists of the above-mentioned coating liquid within limits which follow for example, by which a liquid condition is maintained as an organic EL device is located, the stress relaxation layer made into the purpose can be easily formed by performing formation of a solid-state layer, and formation of an adhesives layer one by one.

[0021] In addition, as for coating liquid, it is desirable also in a liquid with small generating of said shear stress to use as a solvent what evaporates easily under a room temperature. Moreover, since what adhesives permeate does not make business, when it becomes a solid-state layer, it is desirable [the solid-state component which has melted into coating liquid], although there is no need that the aforementioned solid-state layer becomes hard especially that it is what does not let adhesives pass substantially.

[0022] As an example of the coating liquid mentioned above, the thing of following the (1) – (3) is mentioned.

(1) Fluorine system coating liquid, such as what makes a solute what melted Teflon AF (trade name; Du Pont make) to SAITOPPU CTX-105A (trade name; Asahi Glass Co., Ltd. make), a FURORO barrier (trade name; made in the Yasunari company), and FURORINATO FC72 (trade name; product made from Sumitomo 3M), and the thing to which the aforementioned fluorine system liquid was used as the solvent, and the degree of polymerization progressed from that of this liquid.

(2) Silicone system coating liquid, such as what makes a solute that to which the aforementioned silicone system liquid was used as the solvent, and the degree of polymerization progressed from that of this liquid.

(3) Hydrocarbon system coating liquid, such as what makes a solute that to which the aforementioned hydrocarbon system liquid was used as the solvent, and the degree of polymerization progressed from that of this liquid.

[0023] Moreover, as "matter with small generating of shear stress" mentioned above, grease and gel are also suitable. Although grease consists of base oil, a thickening agent, and an additive, especially the thing that uses the liquid with small generating of said shear stress for base oil is desirable. As a thickening agent and an additive, when it is a solid particulate, the one where the particle size is smaller is desirable. If the aforementioned particle size is too large, an organic EL device will be damaged and that will become a short cause. Moreover, the aforementioned particle size needs to be smaller than the thickness made into the purpose of a stress relaxation layer at least. However, if the aforementioned particle size is too small, whenever [butterfly] will be lost, and the advantage using grease is lost. The aforementioned particle size has 100A – desirable 10 micrometers. Since the grease using what is not a solid particulate as a thickening agent and an additive does not have a fear of damaging an organic EL device with the grease concerned, it is much more desirable.

[0024] As an example of the above-mentioned grease, the thing of following the (1) – (3) is mentioned.

(1) Fluorine system grease, such as PTFE grease (trade name; NICHIAS CORP. make).

(2) Silicone system grease, such as grease for FS high vacuums (trade name; Dow Corning make).

(3) Hydrocarbon system grease, such as Apiezon grease N (trade name; the Apiezon company make).

[0025] And as an example of the aforementioned gel, a glycerol, gelatin, polyacrylamide gel, agarose gel, methyl cellulose gel, etc. are mentioned. The gel used by this invention needs to have the fluidity.

[0026] Although formation of a stress relaxation layer may be performed by the approach mentioned above when using the coating liquid which was mentioned above in forming a stress relaxation layer, you may carry out as follows using an accessory plate. First, on an organic EL device, the coating liquid of sufficient amount for a wrap is dropped thru/or applied for this organic EL device, and the layer of coating liquid is once formed. Next, an accessory plate is arranged on the aforementioned layer and the aforementioned layer is uniformly spread the weight of this accessory plate, or by applying a load to this accessory plate. When plane view is carried out as the aforementioned accessory plate, what has sufficient magnitude for a wrap for an organic EL device is used, and this accessory plate is formed in the substrate with which the organic EL device is formed, and the sense which counters substantially. Moreover, the load added to the amount of coating liquid, the weight of an accessory plate, or an accessory plate is suitably adjusted so that the layer of coating liquid may be formed between an organic EL device and an accessory plate. Subsequently, a solid-state layer (coat) is made to form in the front face of a direction parallel to a substrate front face in the outside surface of the layer of coating liquid (field which is in contact with the open air), i.e., the aforementioned layer, by carrying out suitable time amount neglect. At this time, neglect time amount is suitably adjusted so that coating liquid may not solidify about the periphery of an organic EL device. Although a stress relaxation layer is obtained once by this, since the coating liquid of the periphery of an organic EL device solidifies to the inside of a short time the way things stand, an adhesives layer is formed as the aforementioned accessory

plate is covered. Since the vaporization of the solvent in coating liquid is inhibited after forming an adhesives layer, solidification of coating liquid is inhibited, and the coating liquid which exists in the periphery of an organic EL device as the result continues maintaining a liquid condition. The stress relaxation layer made into the purpose by this is obtained.

[0027] Moreover, formation of the stress relaxation layer which uses the grease mentioned above can be performed by dropping thru/or applying the grease of sufficient amount for a wrap for an organic EL device on an organic EL device, and forming a grease layer. Or after forming a grease layer as mentioned above, an accessory plate can be arranged on this grease layer, and it can carry out by spreading the aforementioned grease layer uniformly the weight of this accessory plate, or by applying a load to this accessory plate. Moreover, although it is also possible to form a stress relaxation layer by applying grease to mum order, without using an accessory plate, plastering thinly to mum order may damage an organic EL device, and it can be said to be difficult. And formation of the stress relaxation layer which uses the gel mentioned above can be performed like the case where a stress relaxation layer is formed using grease.

[0028] The thickness (thickness on an organic EL device) of the stress relaxation layer which can be formed as mentioned above is selectable suitably within the limits of 0.1 micrometers – 1cm in general. It is difficult for thickness to acquire desired effectiveness in less than 0.1 micrometers. On the other hand, if thickness exceeds 1cm, since the advantage of an organic electroluminescence device called a thin shape is not only spoiled, but the increase of the volume by thermal expansion will become large according to increase of the volume of a stress relaxation layer, a possibility of destroying by the temperature change at the time of use etc. becomes strong. The thickness with a desirable stress relaxation layer is 0.01–1mm. In addition, thickness here means the thickness of the layer of the coating liquid which exists maintaining a liquid condition on the periphery of an organic EL device when forming using the coating liquid which the stress relaxation layer mentioned above.

[0029] Moreover, although this accessory plate may have the closure effectiveness and does not need to have it when using an accessory plate in forming a stress relaxation layer, it is more more desirable to have the closure effectiveness. It is desirable to use the tabular thing made as an accessory plate by resin, such as inorganic substances, such as glass, ceramics, and a metal, and fluororesin, acrylic resin, a polycarbonate, polyester, a polyamide, polystyrene, polypropylene, polyolefine system resin, etc. As mentioned above, when plane view of the magnitude of an accessory plate is carried out, it is desirable that it is sufficient magnitude for a wrap about an organic EL device. Moreover, as for the thickness of an accessory plate, it is desirable to be referred to in general as 0.1 micrometers – 3mm. The production cost of thickness is high, and a less than 0.1-micrometer thing does not have it from being hard to deal with it. [desirable] On the other hand, if thickness exceeds 3mm, the advantage of an organic electroluminescence device called a thin shape will be spoiled. The desirable thickness of an accessory plate is 0.01–1mm.

[0030] In the organic electroluminescence device of this invention, the stress relaxation layer which mentioned it above as covered the organic EL device formed on the substrate is prepared, and as the organic EL device covered with this stress relaxation layer is covered, the adhesives layer is formed on the aforementioned substrate. While it is for making the sealing agent mentioned later fix, this adhesives layer is for preventing that the stress relaxation layer mentioned above touches with the open air, and unless the configuration of the sealing agent which mentions later whether that side face prepares a special member is made special, it meets with the open air. Therefore, an adhesives layer has especially the desirable thing what has the closure effectiveness, i.e., water permeability, and oxygen permeability are [a thing] low. What consists of photoresist adhesives, thermosetting adhesive, anaerobic adhesive, etc. as such an adhesives layer is desirable, and BENEFIKKUSU VL (trade name of the photoresist adhesives Made from ADERU), acrylic one #4111 (trade name of the photoresist adhesives by Maruto Co., Ltd.), the ARON tightness UL (trade name of the anaerobic adhesive made from Toagosei Chemical industry), the AREMUKO bond 570 (trade name of the thermosetting adhesive Made from ODEKKU), etc. are mentioned as an example.

[0031] An adhesives layer trickles adhesives into the request part of a part in which the aforementioned stress relaxation layer is not prepared in the substrate front face of the side which prepared the stress relaxation layer top (on [when an accessory plate is used in forming a stress relaxation layer] this accessory plate) mentioned above, and this stress relaxation layer. The sealing agent besides mentioned later is arranged, and after spreading the aforementioned adhesives uniformly the weight of a sealing agent, or by applying a load to a sealing agent, it can form by stiffening the adhesives concerned. Moreover, after forming a non-hardened adhesives layer by approaches, such as the applying method, a spin coat method, and a dip method, and arranging the sealing agent later mentioned on this adhesives layer, it can form also by stiffening the adhesives concerned.

[0032] Final thickness of an adhesives layer (thickness on a stress relaxation layer.) When an accessory plate is used in forming a stress relaxation layer, as for the thickness on this accessory plate, it is desirable to be referred to in general as 0.1 micrometers – 1cm. When thickness carries a sealing agent in less than 0.1 micrometers, as a result of the sealing agent concerned coming floating with the surface tension of the adhesives in the condition of not hardening, air bubbles tend to go into the inside of the adhesives in the condition of not hardening, or the interface of this adhesives and sealing agent. On the other hand, if thickness exceeds 1cm, the advantage of an organic electroluminescence device called a thin shape will be spoiled. The thickness with a desirable adhesives layer is 0.001–1mm. Since the organic EL device is covered with the stress relaxation layer at the time of formation of an adhesives layer, there is no need of paying attention in the degree of hardness and contraction at the time of hardening of adhesives substantially. As the result, the width of face of the selection of adhesives from the case where there is no stress relaxation layer spreads.

[0033] Since the sealing agent which is by the adhesives layer mentioned above on a substrate (that in which the organic EL device is formed) is for moisture and oxygen to prevent invasion from the exterior to an organic EL device, it is desirable to use the high thing of the closure effectiveness, i.e., a permeable and oxygen penetrable small thing. As an example of the matter which can be used as a sealing agent, resin, such as inorganic substances, such as glass, ceramics, and a metal, and fluororesin, acrylic resin, a polycarbonate, polyester, a polyamide, polystyrene, polypropylene, polyolefine system resin, is mentioned.

[0034] Since the time and effort of processing is economical few, as for the configuration of a sealing agent, it is desirable that it is tabular. Moreover, although the thinner one of the thickness of a sealing agent is [from / in obtaining a thin organic electroluminescence device] desirable, since it will become weak to impacts, such as external force, if too thin, it is desirable to choose suitably within the limits of 0.1 micrometers – 1cm in general according to the quality of the material of a sealing agent. It is difficult to obtain that in which thickness has practically sufficient resistance to impacts, such as external force, in less than 0.1 micrometers. On the other hand, if thickness exceeds 1cm, it will become difficult to obtain a thin organic electroluminescence device. The more desirable thickness of a sealing agent is 0.01–1mm.

[0035] The organic electroluminescence device of this invention makes the organic EL device covered with the stress relaxation layer mentioned above, the adhesives layer, and the aforementioned stress relaxation layer other than a sealing agent the requirements for an indispensable configuration. That an organic EL device should just be formed on a substrate, the lamination will not be limited, especially if it functions as an organic EL device.

[0036] As an example of the lamination of the organic EL device of the type which makes a substrate side the ejection side of light, the thing of following the (1) – (4) is mentioned for the order of a laminating on a substrate front face.

(1) An anode plate / luminous layer / cathode (2) anode plate / luminous layer / electron injection layer / cathode (3) anode plate / hole-injection layer / luminous layer / cathode (4) anode plate / hole-injection layer / luminous layer / electron injection layer / cathode [0037] Here, although a luminous layer is usually formed of one sort or two or more sorts of organic luminescent material, it may be formed with mixture with an organic luminescent material, a hole-injection ingredient, and/or an electron injection ingredient. Moreover, the protective layer for preventing invasion of the moisture to a component, as the component concerned is covered on the periphery of the component of lamination mentioned above may be prepared.

[0038] When making a substrate side into the ejection side of light, the tabular object which the aforementioned substrate consists of matter which gives high permeability (in general 80% or more) to luminescence (EL light) from an organic EL device at least, and specifically consists of clear glass, a transparent plastic, a quartz, etc., a sheet-like object, or a film-like object is used. In addition, it is also possible to make into the ejection side of light the sealing agent side mentioned above in the organic electroluminescence device of this invention. In this case, it is not necessary to make the order of a laminating on a substrate into above-mentioned sequence and above-mentioned reverse, to form an organic EL device, and to take into consideration the permeability of luminescence (EL light) from an organic EL device about a substrate ingredient. Instead, the matter which gives high permeability (in general 80% or more) to luminescence (EL light) from an organic EL device about the ingredient of a stress relaxation layer, the ingredient of an adhesives layer, and the ingredient of a sealing agent is used.

[0039] As an ingredient of an anode plate, cathode, a luminous layer, a hole-injection layer, an electron injection layer, and a protective layer, a well-known ingredient can be used conventionally, respectively. For example, the metal with a large (for example, 4eV or more) work function as an anode material, an alloy, electrical conductivity compounds, or such mixture are used preferably. As an example, metals, such as gold and nickel, dielectric transparent materials, such as CuI, ITO, SnO₂, and ZnO, etc. are mentioned. Especially, the point of productivity or a controllability to ITO is desirable. Although the thickness of an anode plate is based also on an ingredient, it is usually selectable suitably within the limits of 10nm – 1 micrometer.

[0040] Moreover, as a cathode material, a small (for example, 4eV or less) metal, an alloy, electrical conductivity compounds, or such mixture of a work function etc. are used preferably. As an example, rare earth metals, such as the alloy of a sodium and sodium-potassium alloy, magnesium, a lithium, magnesium, and silver or a mixed metal, aluminum, aluminum/AIO₂, an indium, and an ytterbium, etc. are mentioned. Although the thickness of an anode plate is based also on an ingredient, it is usually selectable suitably within the limits of 10nm – 1 micrometer. Also in any of an anode plate and cathode, below hundreds of ohms / ** of the sheet resistance are desirable. In addition, in case an anode material and a cathode material are chosen, the magnitude of the work function made into criteria is not limited to 4eV.

[0041] The impregnation function in which an electron can be poured in from cathode or an electron injection layer while the ingredient (organic luminescent material) of a luminous layer can pour in an electron hole from an anode plate or a hole-injection layer at the time of the luminous layer for organic EL devices, i.e., electric-field impression. What is necessary is just to be able to form the layer which has the transportation function to which the poured-in charge (an electron and an electron hole at least on the other hand) is moved by the force of electric field, the luminescence function to offer the place of the recombination of an electron and an electron hole and to tie this to luminescence, etc. As the example, the fluorescent brightener of systems, such as a benzothiazole system, a benzimidazole system, and a benzooxazole system, A metal chelation oxy-NOIDO compound, a styryl benzenoid compound, a JISUCHIRIRU pyrazine derivative, Polyphenyl system compound and 12-phtalo peri non, 1, 4-diphenyl-1,3-butadiene, 1, 1, 4, and 4-tetra-phenyl-1,3-butadiene, the North America Free Trade Agreement RUIMIDO derivative, The metal complex of a perylene derivative, an OKISA diazole derivative, an aldazine derivative, a

PIRAJIRIN derivative, a cyclopentadiene derivative, a pyrrolo pyrrole derivative, a styryl derivative, a coumarin system compound, an aromatic dimethylidyne compound, and an eight-quinolinol derivative etc. is mentioned. Although especially the thickness of a luminous layer is not limited, it is usually suitably chosen within the limits of 5nm - 5 micrometers.

[0042] The ingredient (hole-injection ingredient) of a hole-injection layer should just have any of the impregnation nature of an electron hole, and electronic obstruction nature they are. As the example, a triazole derivative, an OKISA diazole derivative, An imidazole derivative, the poly aryl alkane derivative, a pyrazoline derivative, A pyrazolone derivative, a phenylenediamine derivative, an arylamine derivative, An amino permutation chalcone derivative, an oxazole derivative, a styryl anthracene derivative, full — me — non — a derivative, a hydrazone derivative, a stilbene derivative, and a silazane derivative — Conductive polymer oligomer, such as a polysilane system compound, an aniline system copolymer, and thiophene oligomer, a porphyrin compound, an aromatic series tertiary-amine compound, a styryl amine compound, an aromatic series JIMECHIRI DIN system compound, etc. are mentioned. Although especially the thickness of a hole-injection layer is not limited, either, it is usually suitably chosen within the limits of 5nm - 5 micrometers. A hole-injection layer may be 1 layer structure which consists of one sort of the ingredient mentioned above, or two sorts or more, and may be two or more layer structure which consists of two or more layers of the same presentation or a different-species presentation.

[0043] An electron injection layer that what is necessary is just to have the function to transmit the electron poured in from cathode to a luminous layer as an example of the ingredient (electron injection ingredient) nitration full — me — non — a derivative, an anthra quinodimethan derivative, and a diphenyl quinone derivative — Heterocycle tetracarboxylic acid anhydrides, such as a thiopyran dioxide derivative and naphthalene perylene, A carbodiimide, a deflection ORENIRIDEN methane derivative, an anthra quinodimethan derivative, A thing, a JISUCHIRIRU pyrazine derivative, etc. with which the metal complex of an anthrone derivative, an OKISA diazole derivative, and an eight-quinolinol derivative, a metal free phthalocyanine, metal phthalocyanines, or these ends are permuted by the alkyl group, the sulfone radical, etc. are mentioned. Although especially the thickness of an electron injection layer is not limited, either, it is usually suitably chosen within the limits of 5nm - 5 micrometers. An electron injection layer may be 1 layer structure which consists of one sort of the ingredient mentioned above, or two sorts or more, and may be two or more layer structure which consists of two or more layers of the same presentation or a different-species presentation.

[0044] And the copolymer which is made to carry out copolymerization of the monomer mixture containing tetrafluoroethylene and at least one sort of comonomers as an example of the ingredient of a protective layer, and is obtained, The fluorine-containing copolymer which has cyclic structure in a copolymerization principal chain, polyethylene, polypropylene, Polymethylmethacrylate, polyimide, polyurea, polytetrafluoroethylene, Polychlorotrifluoroethylene resin, poly dichlorodifluoroethene, The copolymer of chlorotrifluoroethylene and dichlorodifluoroethene, The absorptivity matter of 1% or more of water absorption, and the dampproof matter of 0.1% or less of water absorption, Metals, such as In, Sn, Pb, Au, Cu, Ag, aluminum, Ti, and nickel, MgO, SiO, SiO₂, aluminum 2O₃, GeO, NiO, CaO, BaO and Fe 2O₃, Y₂O₃, and TiO₂ etc. — a metallic oxide, MgF₂, LiF, AlF₃, and CaF₂ etc. — a metal fluoride etc. is mentioned.

[0045] Moreover, it is not limited especially about the formation approach of each class (an anode plate and cathode are included) which constitutes an organic EL device, either. as the formation approach of an anode plate, cathode, a luminous layer, a hole-injection layer, and an electron injection layer — a vacuum deposition method, a spin coat method, the cast method, the sputtering method, and LB — although law etc. is applicable, about a luminous layer, it is desirable to apply approaches other than the sputtering method (LB a vacuum deposition method, a spin coat method, the cast method, law, etc.). As for especially a luminous layer, it is desirable that it is the molecule deposition film. the thing of the thin film which deposition was carried out to the molecule deposition film from the ingredient compound of a gaseous-phase condition here, and was formed, and the film solidified and formed from the ingredient compound of a solution condition or a liquid phase condition — it is — usually — this molecule deposition film — LB — with the thin film (molecule built up film) formed of law, it is classifiable with the difference of condensation structure and higher order structure, and the functional difference resulting from it. In forming a luminous layer with a spin coat method etc., it prepares a coating solution by melting a binder and ingredient compounds, such as resin, to a solvent.

[0046] moreover — a protective layer — a vacuum deposition method, a spin coat method, the sputtering method, the cast method, and MBE (molecular beam epitaxy) — law, the ionized cluster beam method, the ion plating method, a plasma polymerization method (the high-frequency excitation ion plating method), a reactive-sputtering method, a plasma-CVD method, a laser CVD method, a heat CVD method, a gas source CVD method, etc. are applicable.

[0047] The formation approach of each class can be suitably changed according to the ingredient to be used. If a vacuum deposition method is used in formation of each class which constitutes an organic EL device, since an organic EL device can be formed only with this vacuum deposition method, it is advantageous when aiming at simplification of a facility, and compaction of the production time.

[0048] In addition, the configuration and magnitude of each class (a protective layer is included) which constitute an organic EL device are not necessarily the same. Moreover, when plane view of the component is carried out, all other layers are not surely settled on the electrode formed in right above [of a substrate]. On the other hand, the stress relaxation layer as used in the field of this invention is for controlling that the residual stress of an adhesives layer spreads to an organic EL device, and short-circuit arises in the organic EL device concerned. Therefore, "it is

a wrap stress relaxation layer about "organic EL device" as used in the field of this invention means the stress relaxation layer which has covered at least the field with which cathode and an anode plate lap, when plane view of the organic EL device is carried out. Moreover, "it is the coating liquid (grease, gel) of sufficient amount for a wrap about an organic EL device" as used in this specification means "it is the coating liquid (grease, gel) of amount sufficient at least for a wrap about the field with which cathode and an anode plate lap when plane view of the organic EL device is carried out." And "the accessory plate which has sufficient magnitude for a wrap for an organic EL device when plane view is carried out" means "the accessory plate which has magnitude sufficient at least for a wrap for the field with which cathode and an anode plate lap when plane view is carried out, and plane view of the organic EL device is carried out."

[0049] Moreover, the number of the organic EL devices which constitute the organic electroluminescence device of this invention may be one, and may be plurality. When two or more, each component may have the same class seen from lamination or the luminescent color, and may differ. It is selectable suitably into how many the number of organic EL devices is made according to the application of the organic electroluminescence device made into the purpose etc. When two or more organic EL devices are formed on the substrate, a stress relaxation layer may be prepared for every organic EL device, one thing common to all organic EL devices may be prepared, and two or more things common to the plurality of all the organic EL devices may be prepared. In forming two or more organic EL devices formed on the substrate in a wrap sake for two or more stress relaxation layers, the same is said of the case where an accessory plate is used, and one thing common to [may use an accessory plate for every stress relaxation layer, and] all stress relaxation layers which it is going to prepare may be prepared, and two or more things common to the plurality of all the stress relaxation layers may be prepared. About an adhesives layer and a sealing agent, even when two or more organic EL devices are formed on the substrate, it is desirable practically to prepare one thing common to all organic EL devices, respectively.

[0050] the organic EL device which mentioned above the organic electroluminescence device of this invention, and this organic EL device — a wrap — the above-mentioned stress relaxation layer which made it like and was formed on the substrate (that in which the organic EL device is formed), and this stress relaxation layer — a wrap — it has the above-mentioned adhesives layer which made it like and was formed on the aforementioned substrate, and the above-mentioned sealing agent which fixed on the aforementioned substrate by this adhesives layer. In this organic electroluminescence device, since the organic EL device is covered with the stress relaxation layer, it is controlled that the residual stress of an adhesives layer spreads to an organic EL device. Since it is prevented that the crush of the edge of an organic EL device etc. is carried out by the residual stress spread from the adhesives layer, and it short-circuits with it as the result, the luminescence function of an organic EL device continues at a long period of time, and it is hard to lose it.

[0051] Moreover, although the force works in the parallel direction with the front face of an organic EL device as usual in the organic electroluminescence device of this invention at the time of formation of an adhesives layer, it is controlled from the organic EL device being covered with the stress relaxation layer that the aforementioned force spreads to an organic EL device. Since it is prevented by the organic EL device according to the aforementioned force as the result that electrode exfoliation occurs, it cannot break out easily that the luminescence function of an organic EL device loses after the manufacture process of an organic electroluminescence device or manufacture. Furthermore, since the organic electroluminescence device of this invention closes an organic EL device by the adhesion type closure approach, its closure effectiveness of an organic EL device is high, and its thickness of the device itself is thin, and it is comparatively easy to mass-produce.
 [0052] The organic electroluminescence device of this invention which has such a description can be used as the surface light source, a character indicating equipment, the equipment for electric spectacles, the indicator for mount, the light source for electric discharge of a copying machine, the light source for printers, light modulation equipment, etc.

[0053] Next, the closure approach of the organic EL device of this invention is explained. This approach is characterized by preparing an adhesives layer in it, as this organic EL device is covered at the tooth back of an organic EL device prepared on the substrate, and forming said organic EL device for a wrap stress relaxation layer between said organic EL devices and said adhesives layers in closing said organic EL device by fixing a sealing agent on said substrate by this adhesives layer, as mentioned above.

[0054] Preparing in order to control that the residual stress of an adhesives layer spreads the aforementioned stress relaxation layer to an organic EL device, the ingredient and the formation approach are as having explained in the organic electroluminescence device of this invention mentioned above. Moreover, it is as having also explained the detail of the closure which fixes the organic EL device covered with this stress relaxation layer, and the organic EL device covered with the aforementioned stress relaxation layer by the adhesives layer prepared as covers, and the aforementioned adhesives layer on a substrate (that in which the organic EL device is formed) in the organic electroluminescence device of this invention mentioned above, respectively.

[0055] In the organic EL device closed by this approach, since the organic EL device concerned is covered with the stress relaxation layer, it is controlled that the residual stress of an adhesives layer spreads to an organic EL device. Since it is prevented that the crush of the edge of an organic EL device etc. is carried out by the residual stress spread from the adhesives layer, and it short-circuits with it as the result, the organic EL device (that by which the closure was carried out (= organic electroluminescence device)) which a luminescence function continues at a long period of time, and cannot lose easily is obtained.

[0056] Moreover, although the force works in the direction parallel to the front face of an organic EL device at the time of formation of an adhesives layer, it is controlled from the organic EL device being covered with the stress

relaxation layer that the aforementioned force spreads to an organic EL device. Since it is prevented by the organic EL device according to the aforementioned force as the result that electrode exfoliation occurs, it cannot break out easily that the luminescence function of an organic EL device loses after the process of the closure or the closure. Furthermore, since the approach of this invention is one of the adhesion type closure approaches, it is comparatively easy for the closure effectiveness to be high and to mass-produce an organic EL device (that by which the closure was carried out (= organic electroluminescence device)) with thin thickness.

[0057]

[Example] Hereafter, the example of this invention is explained.

After preparing what formed the ITO film of 100nm of thickness to band-like at the longitudinal direction of the glass substrate concerned (this is hereafter called transparency support substrate) on production place ** of an example 1 (1) organic EL device, and the glass substrate of 25mmx75mmx1.1mm magnitude and cleaning this transparency support substrate ultrasonically for 30 minutes by isopropyl alcohol, pure water washed for 30 minutes and, finally it cleaned ultrasonically for 30 minutes by isopropyl alcohol again. The transparency support substrate after washing is fixed to the substrate electrode holder of a commercial vacuum evaporation system (product made from Japanese Vacuum technology). On the resistance heating boat made from molybdenum, N, N'-diphenyl-N, N'-screw-(3-methylphenyl)-[1 and 1'-biphenyl]-4, and 4'-diamine It put in (it having been hereafter called TPD), 200mg (henceforth Alq3) of tris (eight quinolinol) aluminum was put into another resistance heating boat made from molybdenum, and the inside of a vacuum chamber was decompressed up to 1x10 to 4 Pa. [200mg]

[0058] Next, heated the aforementioned resistance heating boat into which TPD was put to 215-220 degrees C. TPD was made to deposit on the ITO film in the evaporation rate of 0.1-0.3nm/second, and the hole-injection layer of 60nm of thickness was formed. The substrate temperature at this time was a room temperature. Subsequently, the luminous layer was successively formed to membrane formation of a hole-injection layer, without taking out the transparency support substrate with which the hole-injection layer was formed from a vacuum chamber. Membrane formation of a luminous layer is Alq3. The put-in aforementioned resistance heating boat is heated to 275 degrees C, and it is Alq3. It is made to deposit on a hole-injection layer in the evaporation rate of 0.1-0.2nm/second, and is Alq3 of 60nm of thickness. It carried out by forming a layer. The substrate temperature at this time was also a room temperature. Next, magnesium 1g was put into the resistance heating boat made from molybdenum, 500mg of silver was put into another resistance heating boat made from molybdenum, and the inside of a vacuum chamber was decompressed up to 2x10 to 4 Pa. And while heating the aforementioned resistance heating boat into which magnesium was put at about 500 degrees C and evaporating magnesium in the evaporation rate of about 1.7-2.8nm/second, the aforementioned resistance heating boat into which silver was put was heated at about 800 degrees C, silver was evaporated in the evaporation rate of 0.03-0.08nm/second, and the cathode of 150nm of thickness which consists of a mixed metal of magnesium and silver was established in a total of on [three] a luminous layer. The magnitude on plane view of each cathode is 3mmx15mm, and these are prepared in the sense which intersects perpendicularly with the ITO film at fixed spacing.

[0059] Then, the glass substrate prepared to the aforementioned cathode was cut to three, and a total of three organic EL devices whose lamination on a glass substrate is an anode plate (ITO film) / hole-injection layer / luminous layer / cathode (Mg-Ag layer) were obtained. The magnitude of the luminescence side (field where cathode and an anode plate lap in when plane view is carried out, and the hole-injection layer and the luminous layer exist among these electrodes) of these organic EL devices is 3mmx5mm. Moreover, the initial brightness of these organic EL devices is electrical-potential-difference 6.5V and current density 3 mA/cm². They are 100 cd/m² under conditions. Reaching, the power conversion effectiveness at this time was 1.6 lm/W.

[0060] (2) the formation list of a stress relaxation layer and an adhesives layer — as the ingredient of arrangement place ** of a sealing agent, and a stress relaxation layer — fluororesin coating liquid (SAITOPPU CTX-105A by Asahi Glass Co., Ltd.) — preparing — moreover — as an accessory plate — about 10 — the cover glass (henceforth cover glass I) of mmx10mmx0.15mm magnitude was prepared. Next, it was dropped on the organic EL device which produced the 5micro of the aforementioned fluororesin coating liquid I above (1), and the aforementioned cover glass I was carried on this. The thin film (0.05mm in thickness) of said coating liquid was uniformly made by said coating liquid with the viscous lowness of fluororesin coating liquid, and the weight of cover glass I between breadth, an organic EL device, and cover glass I. At this time, some coating liquid was protruded into the outside of the field covered by cover glass I a little. By leaving it about 10 minutes in this condition, the solid-state layer (coat) was made to form in the outside surface of the above-mentioned thin film which consists of fluororesin coating liquid, i.e., the front face of a glass substrate and the front face of an parallel direction, and the stress relaxation layer made into the purpose was obtained once. At this time, the fluororesin coating liquid of the part which is in contact with the organic EL device was maintaining the liquid condition, without solidifying.

[0061] Next, 0.1ml (BENEFIKKUSU VL Made from ADERU) of photoresist adhesives was dropped on the above-mentioned cover glass I as adhesives, and the cover glass (magnitude is called cover glass II below about 20mmx20mmx0.15mm;) as a sealing agent was carried on it. Said photoresist adhesives covered completely breadth, the above-mentioned stress relaxation layer, and cover glass I uniformly with the viscous lowness of photoresist adhesives, and the weight of cover glass II. Following on installation of cover glass II, the light of a halogen lamp was irradiated from on the aforementioned cover glass II at photoresist adhesives, and the photoresist adhesives concerned were stiffened.

[0062] The adhesives layer with a thickness of 0.1mm (thickness on cover glass I) which consists of photoresist adhesives by this as covers the above-mentioned stress relaxation layer was formed, and the cover glass II as a

sealing agent fixed by the aforementioned adhesives layer to coincidence on the glass substrate (that in which the organic EL device is formed). Furthermore, since the vaporization of the solvent of the fluororesin coating liquid in the above-mentioned stress relaxation layer was inhibited after forming an adhesives layer, finally the stress relaxation layer to which it continues maintaining the fluororesin coating liquid which exists in the periphery of an organic EL device, and it targets a liquid condition as the result was formed.

[0063] By having carried out to formation of an adhesives layer, and arrangement of a sealing agent as mentioned above, the organic electroluminescence device made into the purpose was obtained. Hereafter, the organic electroluminescence device was produced similarly and a total of 45 organic electroluminescence devices were obtained. The schematic diagram of the cross section of the organic electroluminescence device produced by this example 1 is shown in drawing 1 R> 1, and a plan is shown in drawing 2.

[0064] The organic EL device 3 with which this organic electroluminescence device 1 was formed in one side of a glass substrate 2 as shown in drawing 1. The stress relaxation layer 4 formed on the glass substrate 2 as covered this organic EL device 3. The accessory plate 5 which consists of cover glass I used in order to form this stress relaxation layer 4. It has the adhesives layer 6 which consists of photoresist adhesives formed on the glass substrate 2 as covered this accessory plate 5 and said stress relaxation layer 4, and the sealing agent 7 which consists of cover glass II which fixed on the glass substrate 2 by this adhesives layer 6. The stress relaxation layer 4 is made from fluororesin coating liquid, in this stress relaxation layer 4, the front face of a glass substrate 2 and the front face of an parallel direction are solidified, when the solvent evaporated at the time of that formation, solid-state layer 4a is formed, and fluororesin coating liquid 4b of the part which is in contact with the organic EL device 3 is maintaining the liquid condition, without solidifying.

[0065] Moreover, an organic EL device 3 is Alq3 as the ITO film as an anode plate, the TPD layer as a hole-injection layer, and a luminous layer on a glass substrate 2. The laminating of a layer and the Mg—Ag mixolimnion as cathode is carried out one by one. It is sign 3a about the ITO film among these, and sign 3b shows a Mg—Ag mixolimnion in drawing 1 and drawing 2, respectively. As shown in drawing 1 and drawing 2, ITO film 3a is formed in glass substrate 2 front face band-like, and Mg—Ag mixolimnion 3b is prepared in the sense which intersects perpendicularly with ITO film 3a. And the part with which ITO film 3a and Mg—In mixolimnion 3b have lapped on plane view is equivalent to the luminescence side of an organic EL device 3.

[0066] an example 2 — the organic EL device was first produced on the glass substrate on an example 1 and these conditions. next, the this top after applying these fluorine system grease about 0.01ml on the above-mentioned organic EL device, using fluorine system grease (PTFE grease by NICHIAS CORP.) as an ingredient of a stress relaxation layer — as an accessory plate — about 10 — the cover glass I of mmx10mmx0.15mm magnitude was carried. Subsequently, by pressing the aforementioned cover glass lightly from a top, the aforementioned fluorine system grease was spread uniformly and the thin film (0.1mm in thickness) of fluorine system grease was formed between an organic EL device and cover glass I. Thereby, the stress relaxation layer was obtained. Then, formation of the adhesives layer (0.06mm in thickness) which consists of photoresist adhesives on an example 1 and these conditions, and fixing of the sealing agent (cover glass II) by this adhesives layer were performed, and the organic electroluminescence device made into the purpose was obtained. Hereafter, the organic electroluminescence device was produced similarly and a total of 55 organic electroluminescence devices were obtained.

[0067] an example 3 — the organic EL device was first produced on the glass substrate on an example 1 and these conditions. Next, the aluminum layer of 0.2 micrometers of thickness was formed with the vacuum deposition method as a protective layer on the cathode (Mg—Ag layer) which constitutes the aforementioned organic EL device. Then, formation of the stress relaxation layer (0.05mm in thickness) using an accessory plate (cover glass I), formation of the adhesives layer (0.1mm in thickness) which consists of photoresist adhesives, and fixing of the sealing agent (cover glass II) by this adhesives layer were performed on an example 1 and these conditions, and the organic electroluminescence device made into the purpose was obtained. Hereafter, the organic electroluminescence device was produced similarly and a total of 45 organic electroluminescence devices were obtained.

[0068] A total of 45 organic electroluminescence devices were produced like the example 1 except having not formed an example of comparison 1 stress-relaxation layer. An accessory plate was not used at this time, either.

[0069] A total of 45 organic electroluminescence devices were produced like the example 1 except both having not fixed the sealing agent by formation of an example of comparison 2 adhesives layer, and the adhesives layer.

[0070] the example 3 of a comparison — after washing this transparency support substrate on an example 1 and these conditions first, using that by which the 10mmx75mmx100nm ITO film was formed by the edge of both directions of a short hand of one side of the glass substrate of 25mmx75mmx1.1mm magnitude, respectively as a transparency support substrate, the substrate electrode holder of the vacuum evaporation system equipped with the mask automatic switchboard style was equipped. Next, the mask was covered over one side of the above-mentioned ITO film, and the hole-injection layer and the luminous layer were formed on an example 1 and these conditions by this condition, respectively. Subsequently, after removing the above-mentioned mask using the mask automatic switchboard style attached to vacuum evaporation equipment, width of face of 5mm was covered at the radial-border section of both longitudinal directions of one ITO film (ITO film of the direction over which the aforementioned mask was covered), respectively as it is also at said device, and the mask was covered.

[0071] Next, heat the resistance heating boat made from molybdenum into which magnesium 1g was put beforehand, and the resistance heating boat made from molybdenum into which 500mg of silver was put beforehand, respectively, made magnesium vapor-deposit with the evaporation rate of about 1.5nm/second, coincidence was

made to vapor-deposit silver with the evaporation rate of about 0.1nm/second, and the electrode of 150nm of thickness which consists of a mixed metal of magnesium and silver was formed on the luminous layer. By having formed membranes to cathode, the organic EL device was formed on the above-mentioned glass substrate.

[0072] Then, the protective layer was formed in the way of the following [the bottom of a series of vacuum environments which continue from membrane formation of a hole-injection layer, a luminous layer, and cathode], using the above-mentioned vacuum evaporation system as it is. First, the crucible made from an alumina which held 1.5g (Du Pont Teflon AF) of amorphism copolymer powder of tetrafluoroethylene, and perfluoro -2, the 2-dimethyl -1 and 3-JIOKI SOL as an evaporation source is beforehand put into the basket made from a tungsten, and the mesh made from stainless steel of 12 micrometerphi was put above this basket made from a tungsten (on the crucible made from an alumina). Subsequently, after decompressing the inside of a vacuum chamber up to 1x10 to 4 Pa, energization heating was carried out at the aforementioned basket made from a tungsten, the aforementioned evaporation source was heated at 455 degrees C, and the protective layer (Teflon AF thin film) of 800nm of thickness was formed with the evaporation rate of 0.5nm/second on the above-mentioned cathode which constitutes an organic EL device.

[0073] Then, the organic EL device which formed membranes to the protective layer was taken out from the vacuum chamber, the casing type closure was performed in the following ways about this organic EL device, and the organic electroluminescence device was obtained. First, the hole-injection layer which covers width of face of 5mm, respectively, and is prepared on this ITO film about the radial-border section of both longitudinal directions of the ITO film of the direction over which a mask was not covered in membrane formation of a hole-injection layer, a luminous layer, cathode, and a protective layer were excised. Moreover, also about the edge of both directions of a short hand of a glass substrate, width of face of 5mm was covered, respectively, and a hole-injection layer, a luminous layer, cathode, and a protective layer were excised so that the thickness might become the sum of the thickness of a glass substrate, and the thickness of the ITO film substantially.

[0074] Subsequently, the glass plate (outside **: call it 20mmx75mmx3mm and following shield glass) which has a 18mmx73mmx2mm crevice and a through tube (henceforth an inlet) with a diameter of 2mm prepared in the bottom of this crevice is prepared, and this shield glass and the organic EL device which formed membranes to the protective layer were stuck with epoxy system adhesives (Cemedine super by Cemedine Co., Ltd. 5). After epoxy system adhesives mixed base resin and a curing agent and stirred them 20 times by the spatula, they were applied to the rectangle of about 20x75mm by width of face of 1mm at the edge of the organic EL device which formed membranes to the above-mentioned protective layer. Moreover, into the crevice of shield glass, cathode and a protective layer stuck it, as shield glass and the organic EL device which formed membranes to the above-mentioned protective layer were settled. After lamination and in atmospheric air, it was left for 10 hours and epoxy system adhesives were stiffened.

[0075] Subsequently, the silicone oil (it is called TSK451 by Toshiba Corp. and the liquid for the following closures) which did 8 volume % distribution of the silica gel for moisture absorption (particle size of 50 micrometers) was poured in from the inlet established in shield glass, and the inside of the space formed by the crevice of shield glass and the organic EL device which formed membranes to the above-mentioned protective layer was filled with the liquid for the closures. Then, the glass lid blocked the inlet and the organic electroluminescence device with which the casing type closure was given was obtained. In addition, the glass lid was pasted up on shield glass with the epoxy system adhesives mentioned above. Hereafter, the organic electroluminescence device was produced similarly and a total of 45 organic electroluminescence devices were obtained.

[0076] About the organic electroluminescence device produced, respectively in the evaluation example 1 of the yield - the example 3 and the example 1 of a comparison - the example 3 of a comparison, immediately after device production to initial brightness is 100 cd/m². The continuation drive of the organic EL device was carried out by the becoming direct-current constant current, and the number of the organic EL devices which lost the luminescence function more short at each time one week, one month (for 31 days), and 1000 hours after [immediately after continuation drive initiation and] was counted. In addition, measurement of brightness was performed in atmospheric air using the luminance meter (CS-100 by Minolta Co., Ltd.). This result is shown in Table 1.

[0077]

[Table 1]

表 1

評価に付した デバイスの数	発光機能を喪失した素子の数			
	駆動開始直後	1週間後	1箇月後	1000時間後
実施例 1	45	1	1	1
実施例 2	45	3	3	3
実施例 3	45	4	4	4
比較例 1	45	43	45	45
比較例 2	45	2	8	31
比較例 3	45	1	2	3

[0078] As shown in Table 1, there are few organic EL devices which lost the luminescence function more short in the organic electroluminescence device produced in the example 1 – the example 3, respectively. This shows that the reliable organic electroluminescence device was produced under the high yield. On the other hand, in the example 1 of a comparison which did not prepare a stress relaxation layer, almost all organic EL devices had lost the luminescence function more short at the time immediately after continuation drive initiation, and all the organic EL devices had lost the luminescence function more short after one week. This shows that a stress relaxation layer is useful, when an organic EL device prevents losing the luminescence function therefore short.

[0079] Moreover, in the organic electroluminescence device produced in the example 2 of a comparison, although there were few organic EL devices which therefore lost the luminescence function short at the time immediately after continuation drive initiation, 1000 hours after, all the components had lost the luminescence function more short. And in the organic electroluminescence device produced in the example 3 of a comparison, it is at all the investigation time and is [whether the number of the organic EL devices which therefore lost the luminescence function short is equivalent to an example 1 – an example 3, and] less than [it]. Therefore, it can say that the organic electroluminescence device with the high dependability seen from a viewpoint whether an organic EL device therefore loses that luminescence function short was produced under the high yield, and the organic electroluminescence device of the example 3 of a comparison is excellent in the example 3 of a comparison with this point. However, if the closure effectiveness mentioned later, the difficulty of manufacture, the thickness of an organic electroluminescence device, etc. are taken into consideration, it can be said that the direction of the organic electroluminescence device of an example 1 – an example 3 is excellent.

[0080] The round formal field called the dark spot looked at by the evaluation organic EL device of the closure effectiveness non-emitted light is expanded to the organic EL device, when moisture and oxygen invade. Therefore, it is suitable as an index for evaluating the closure effectiveness to investigate aging of the magnitude about the dark spot of arbitration. Immediately after the continuation drive initiation for the evaluation of the yield mentioned above from such a viewpoint, A photograph of each organic electroluminescence device is taken at each [of one day, one week, one month (for 31 days), and 1000 hours after] time. One device was chosen as arbitration from the organic electroluminescence devices which had not lost the luminescence function at the time 1000 hours after, and it asked for each diameter in a time from the aforementioned photograph paying attention to the dark spot of the arbitration in the organic EL device which constitutes the organic electroluminescence device. This result is shown in Table 2. In addition, since all the organic EL devices had lost the luminescence function more short about the organic electroluminescence device produced in the example 1 of a comparison at the time one week after, one device was chosen as arbitration from the organic electroluminescence devices which had not lost the luminescence function at the time one day after. Moreover, since all the organic EL devices had lost the luminescence function more short about the organic electroluminescence device produced in the example 2 of a comparison at the time 1000 hours after, one device was chosen as arbitration from the organic electroluminescence devices which had not lost the luminescence function at the time one month after.

[0081]

[Table 2]

表 2

	駆動開始直後	1 日 後	1 週 間 後	1 箇 月 後	1000 時間後
実施例 1	1 4	1 7 (21%)	1 8 (29%)	2 1 (50%)	2 3 (64%)
実施例 2	1 1	1 5 (36%)	1 9 (73%)	2 3 (109%)	2 5 (127%)
実施例 3	6	8 (33%)	9 (50%)	1 2 (100%)	1 3 (117%)
比較例 1	1 3	1 6 (23%)	—	—	—
比較例 2	1 7	3 2 (88%)	8 0 (371%)	450 (2550%)	—
比較例 3	1 2	1 5 (25%)	2 5 (108%)	3 0 (150%)	3 3 (175%)

* 表内の数字の単位は μm

* 括弧内の数値は駆動開始直後のダークスポットの直径に対する直径の増大分を百分率で表したものである。

* 実線を付した空欄は、全ての素子が発光機能を喪失したために測定できなかったことを示す。

[0082] As shown in Table 2, also after carrying out a continuation drive for 1000 hours, with the organic electroluminescence device produced in the example 1 – the example 3, respectively, the dark spot is not expanded so much. This shows that the high closure effectiveness is acquired to an organic EL device in each organic electroluminescence device of an example 1 – an example 3. O Sufficient data for the organic electroluminescence device of the example 1 of a comparison to estimate the closure effectiveness on the other hand, since the organic EL device which loses a luminescence function more short succeeded one another were not able to be obtained. Moreover, it turns out that growth of a dark spot is very quick and the organic electroluminescence device of the example 2 of a comparison of the closure to an organic EL device is inadequate. And in the organic electroluminescence device of the example 3 of a comparison, growth of a dark spot is a little quick, and the closure effectiveness over an organic EL device understands that nearby [of an example 1 – an example 3] is low.

[0083]

[Effect of the Invention] As explained above, the organic electroluminescence device of this invention is an organic electroluminescence device which the luminescence function of an organic EL device cannot lose easily, closing an organic EL device by the adhesion type closure method. Moreover, in the organic EL device closed by the approach of this invention, it cannot break out easily that the luminescence function of an organic EL device loses after the closure. Therefore, according to this invention, it becomes possible to continue with a thin shape at a long period of time, and to provide the bottom of the high yield with a stable organic electroluminescence device by the comparatively easy production process.

[0084]

[Translation done.]

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TECHNICAL FIELD

[Industrial Application] This invention relates to the closure approach of the organic electroluminescence device equipped with the organic EL device as a source of luminescence, and an organic EL device, and relates to the adhesion type closure approach for closing the organic electroluminescence device and organic EL device which were equipped with the organic electroluminescence by which the closure was especially carried out by the adhesion type closure approach as a source of luminescence.

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PRIOR ART

[Description of the Prior Art] There are an inorganic EL element and an organic EL device in an EL element, since any EL element is self-luminous, visibility is high, and handling is easy while excelling in shock resistance, since it is a perfect solid-state component. For this reason, the researches and developments and utilization as the pixel of graphic display, the pixel of a television image display device, or the surface light source are advanced. An organic EL device makes the luminous layer which contains an organic luminescent material for an anode plate and cathode among these intervene at least, and carries out a laminating, and the thing of various lamination is developed by current. As an example of lamination, an anode plate / luminous layer / cathode, an anode plate / luminous layer / electron injection layer / cathode, an anode plate / hole-injection layer / luminous layer / cathode, an anode plate / hole-injection layer / luminous layer / electron injection layer / cathode, etc. are mentioned. Although a luminous layer is usually formed of one sort or two or more sorts of organic luminescent material, it may be formed with mixture with an organic luminescent material, a hole-injection ingredient, and/or an electron injection ingredient.

[0003] In many cases, the organic EL device which has such lamination is formed on a substrate. And when taking out EL light from a luminous layer from the aforementioned substrate side, an anode plate is formed in right above [of a substrate], and cathode is formed in right above [of a substrate] when taking out EL light from a luminous layer from the side which formed the organic EL device. An anode plate is formed with the transparency thru/or the translucent thin film which consists of big matter of work functions, such as Au, nickel, and ITO (indium stannic acid ghost). On the other hand, cathode is formed with the thin film which consists of small matter of work functions, such as Yb, Mg, aluminum, and In.

[0004] An organic EL device is a light emitting device of a current drive mold, and uses luminescence produced when the electron and electron hole which were poured into the luminous layer (organic luminescent material) recombine. For this reason, the organic EL device has the advantage that a drive by low battery called 4.5V is possible by making thickness of a luminous layer thin, and a response is also quick, and the advantage that luminescence of high brightness is possible since brightness is proportional to an inrush current. moreover, luminescence of various colors, such as 4 gloss blue, green, yellow, and red, is obtained by changing luminescent material. The research for current and utilization is continued by these advantages.

[0005] By the way, the organic substance, such as an organic luminescent material used for an organic EL device, is weak into moisture, oxygen, etc. Moreover, also in an electrode, in atmospheric air, a property deteriorates rapidly by oxidation. Therefore, in order to obtain a practical organic EL device and an organic electroluminescence device, it is necessary to give the closure. As the closure approach of an organic EL device, it is SiO₂ to the approach (refer to JP,4-137483,A) of forming the poly paraxylene film as a closure layer in the component front face for the closure by the vapor-phase-polymerization method, and the component front face for the closure. Although there is also the approach (refer to JP,4-73886,A) of forming a protective coat, the approach that the closure effectiveness is more high is already developed. The approach is divided roughly into the following two gestalten.

[0006] One is the casing type closure approach and it is the approach of closing by putting in an organic EL device in a case, intercepting with the external world, and being filled up with the predetermined fluid for the closures with the organic EL device in the aforementioned case. Another is the adhesion type closure approach and is the approach of closing by carrying out face bonding of the sealing agents, such as a glass plate, to the tooth back (in view of a substrate side back of a component) of the organic EL device currently formed on the substrate with adhesives.

[0007] According to the casing type closure approach, the degradation can be prevented, without destroying an organic EL device by choosing a thing suitable as a fluid for the closures put in in a case, but on the other hand, a difficulty like following the (1) - (3) is produced.

- (1) Since it makes it indispensable to put an organic EL device into a case, the description of the thin shape which is one of the advantages of an organic EL device is spoiled.
- (2) Since the process which pours in the fluid for the closures is required in the process which machines a case, and a case, it is unsuitable for mass production method.
- (3) Like [when it is going to close the organic EL device of a large number currently formed on the substrate using one case], when a lot of liquids for the closures are put into a big case, and the volume of the fluid for the closures within a case increases by the thermal expansion accompanying the temperature rise of the external world, there is a possibility of destroying a case.

[0008] On the other hand, according to the adhesion type closure approach, an organic electroluminescence device (what is equipped with the organic EL device by which the closure was carried out as a source of luminescence)

with comparatively easy mass production method can be easily obtained thinly [the thickness] after the closure, and the high closure effectiveness can be acquired easily. They are direct or SiO₂ to the periphery of the approach (refer to JP,4-212284,A) of fixing a glass substrate on this, after preparing the protective coat which consists of inorganic compounds, such as GeO, in the periphery of the organic EL device for the closure as the adhesion type closure approach, and the organic EL device for the closure. A photo-setting resin layer is prepared through the film, and the approach (refer to JP,5-182759,A) of fixing a glass substrate through this photo-setting resin layer is learned.

[Translation done.]

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EFFECT OF THE INVENTION

[Effect of the Invention] As explained above, the organic electroluminescence device of this invention is an organic electroluminescence device which the luminescence function of an organic EL device cannot lose easily, closing an organic EL device by the adhesion type closure method. Moreover, in the organic EL device closed by the approach of this invention, it cannot break out easily that the luminescence function of an organic EL device loses after the closure. Therefore, according to this invention, it becomes possible to continue with a thin shape at a long period of time, and to provide the bottom of the high yield with a stable organic electroluminescence device by the comparatively easy production process.

[Translation done.]

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] However, when the organic electroluminescence device which closed and obtained the organic EL device by the closure approach of adhesion type [conventional] was made to carry out a continuation drive for about 1000 hours, there was a difficulty that the luminescence function of an organic EL device is lost in it.

[0010] The 1st purpose of this invention is the organic electroluminescence device equipped with the organic EL device by which the closure was carried out by the adhesion type closure method as a source of luminescence, and is to offer the organic electroluminescence device which the luminescence function of an organic EL device cannot lose easily.

[0011] Moreover, the 2nd purpose of this invention is to offer the closure approach of the organic EL device which the luminescence function of an organic EL device cannot lose easily after the closure.

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MEANS

[Means for Solving the Problem] As a result of this invention persons' adding examination wholeheartedly about the cause of loss of the luminescence function mentioned above, it found out loss of this luminescence function not originating in reduction of the closure effectiveness, and originating in short-circuit occurring in the electrode edge by the side of the adhesives layer of an organic EL device. This point is explained in more detail.

[0013] between the adhesives of the organic EL device and the above for the closure, or photo-setting resins — GeO and SiO₂ etc. — even if there is film, if that thickness is thin, it will get across to organic electroluminescence with mum order. [since the volume contracts in adhesives or a photo-setting resin at the time of hardening residual stress occurs, and] [this residual stress] And since residual stress becomes strong especially in a part with small radius of curvature, the residual stress spread to the organic EL device is concentrated on the edge of the component concerned etc. Consequently, the crush of the electrode edge of a component etc. is carried out, an anode plate and cathode contact, and short-circuit occurs. this invention persons reached this invention, as a result of adding examination further based on the above-mentioned knowledge.

[0014] The organic electroluminescence device of this invention which attains the 1st purpose of the above is an organic electroluminescence device equipped with the organic EL device formed on the substrate, the adhesives layer formed on said substrate as covered this organic EL device, and the sealing agent which fixed on said substrate by this adhesives layer, and is characterized by having a wrap stress relaxation layer for said organic EL device between said organic EL devices and said adhesives layers.

[0015] Moreover, the closure approach of the organic EL device of this invention of attaining the 2nd purpose of the above is characterized by preparing an adhesives layer in it, as this organic EL device is covered at the tooth back of an organic EL device prepared on the substrate, and forming said organic EL device for a wrap stress relaxation layer between said organic EL devices and said adhesives layers in closing said organic EL device by fixing a sealing agent on said substrate by this adhesives layer.

[0016] Hereafter, this invention is explained to a detail. If the organic electroluminescence device of this invention is explained first, this organic electroluminescence device will be characterized by having a stress relaxation layer in a specific part, as mentioned above. Then, this stress relaxation layer is explained first.

[0017] The above-mentioned stress relaxation layer is for controlling that the residual stress of an adhesives layer spreads to an organic EL device. Therefore, although this stress relaxation layer may have the effectiveness (henceforth the closure effectiveness) of preventing moisture and oxygen invading into an organic EL device from the exterior and does not need to have it, it has the closure effectiveness and the direction is more desirable. Moreover, as for a stress relaxation layer, it is desirable that they are the layer of the monolayer structure which consists only of a layer which generating of shear stress becomes from the small matter, and the layer of two or more layer structure containing the layer which generating of blank stress becomes from the small matter. In making a stress relaxation layer into two or more layer structure, it forms in the periphery of an organic EL device the layer which generating of shear stress becomes from the small matter.

[0018] Here, the above-mentioned "matter with small generating of shear stress" means the fluid (a gas, liquid) which has the property in which the gap is not told substantially, grease, gel, etc., when an adhesives layer shifts in the direction parallel to the electrode of an organic EL device according to factors, such as hardening.

[0019] Generating of the above-mentioned shear stress can mention hydrocarbon system liquids, such as silicone system liquids, such as fluorine system liquids, such as gases, such as gaseous helium, nitrogen gas, and argon gas, and chlorofluorocarbon, a perfluoro polyether, a perfluoro amine, a perfluoro alkane, methyl hydrogen silicone, methyl chlorophenyl silicone, and trifluoro propylmethyl silicone, an alpha olefin (Pori alpha olefin of C5 – C17), polybutene, alkylbenzene, and a polyalkylene glycol, etc. as a fluid among small matter. However, a complicated process is needed for forming, preventing that the adhesives for adhesives stratification advance the stress relaxation layer which consists of these fluids into the inside of the stress relaxation layer concerned, or the interface of the stress relaxation layer and a substrate (substrate for forming an organic EL device) concerned. Therefore, when using a fluid as the above-mentioned "matter with small generating of shear stress", it is desirable to use the coating liquid which is made to come to dissolve resin, rubber, etc. in a solvent as this fluid, and carries out film formation by evaporation of a solvent under about 10–30-degree C temperature conditions among atmospheric air.

[0020] When the above-mentioned coating liquid is used, it is possible to be able to make a solid-state layer (coat) form in a layer front face, and to maintain a liquid condition about the part inside the aforementioned solid-state layer by evaporating a little the solvent in the layer which consists of the coating liquid concerned. After forming the layer which consists of the above-mentioned coating liquid within limits which follow for example, by which a liquid

condition is maintained as an organic EL device is located, the stress relaxation layer made into the purpose can be easily formed by performing formation of a solid-state layer, and formation of an adhesive layer one by one. [0021] In addition, as for coating liquid, it is desirable also in a liquid with small generating of said shear stress to use as a solvent what evaporates easily under a room temperature. Moreover, since what adhesives permeate does not make business, when it becomes a solid-state layer, it is desirable [the solid-state component which has melted into coating liquid], although there is no need that the aforementioned solid-state layer becomes hard especially that it is what does not let adhesives pass substantially.

[0022] As an example of the coating liquid mentioned above, the thing of following the (1) – (3) is mentioned.

(1) Fluorine system coating liquid, such as what makes a solute what melted Teflon AF (trade name; Du Pont make) to SAITOPPU CTX-105A (trade name; Asahi Glass Co., Ltd. make), a FURORO barrier (trade name; made in the Yasunari company), and FURORINATO FC72 (trade name; product made from Sumitomo 3M), and the thing to which the aforementioned fluorine system liquid was used as the solvent, and the degree of polymerization progressed from that of this liquid.

(2) Silicone system coating liquid, such as what makes a solute that to which the aforementioned silicone system liquid was used as the solvent, and the degree of polymerization progressed from that of this liquid.

(3) Hydrocarbon system coating liquid, such as what makes a solute that to which the aforementioned hydrocarbon system liquid was used as the solvent, and the degree of polymerization progressed from that of this liquid.

[0023] Moreover, as "matter with small generating of shear stress" mentioned above, grease and gel are also suitable. Although grease consists of base oil, a thickening agent, and an additive, especially the thing that uses the liquid with small generating of said shear stress for base oil is desirable. As a thickening agent and an additive, when it is a solid particulate, the one where the particle size is smaller is desirable. If the aforementioned particle size is too large, an organic EL device will be damaged and that will become a short cause. Moreover, the aforementioned particle size needs to be smaller than the thickness made into the purpose of a stress relaxation layer at least. However, if the aforementioned particle size is too small, whenever [butterfly] will be lost, and the advantage using grease is lost. The aforementioned particle size has 100A – desirable 10 micrometers. Since the grease using what is not a solid particulate as a thickening agent and an additive does not have a fear of damaging an organic EL device with the grease concerned, it is much more desirable.

[0024] As an example of the above-mentioned grease, the thing of following the (1) – (3) is mentioned.

(1) Fluorine system grease, such as PTFE grease (trade name; NICHIAS CORP. make).

(2) Silicone system grease, such as grease for FS high vacuums (trade name; Dow Corning make).

(3) Hydrocarbon system grease, such as Apiezon grease N (trade name; the Apiezon company make).

[0025] And as an example of the aforementioned gel, a glycerol, gelatin, polyacrylamide gel, agarose gel, methyl cellulose gel, etc. are mentioned. The gel used by this invention needs to have the fluidity.

[0026] Although formation of a stress relaxation layer may be performed by the approach mentioned above when using the coating liquid which was mentioned above in forming a stress relaxation layer, you may carry out as follows using an accessory plate. First, on an organic EL device, the coating liquid of sufficient amount for a wrap is dropped thru/or applied for this organic EL device, and the layer of coating liquid is once formed. Next, an accessory plate is arranged on the aforementioned layer and the aforementioned layer is uniformly spread the weight of this accessory plate, or by applying a load to this accessory plate. When plane view is carried out as the aforementioned accessory plate, what has sufficient magnitude for a wrap for an organic EL device is used, and this accessory plate is formed in the substrate with which the organic EL device is formed, and the sense which counters substantially. Moreover, the load added to the amount of coating liquid, the weight of an accessory plate, or an accessory plate is suitably adjusted so that the layer of coating liquid may be formed between an organic EL device and an accessory plate. Subsequently, a solid-state layer (coat) is made to form in the front face of a direction parallel to a substrate front face in the outside surface of the layer of coating liquid (field which is in contact with the open air), i.e., the aforementioned layer, by carrying out suitable time amount neglect. At this time, neglect time amount is suitably adjusted so that coating liquid may not solidify about the periphery of an organic EL device. Although a stress relaxation layer is obtained once by this, since the coating liquid of the periphery of an organic EL device solidifies to the inside of a short time the way things stand, an adhesives layer is formed as the aforementioned accessory plate is covered. Since the vaporization of the solvent in coating liquid is inhibited after forming an adhesives layer, solidification of coating liquid is inhibited, and the coating liquid which exists in the periphery of an organic EL device as the result continues maintaining a liquid condition. The stress relaxation layer made into the purpose by this is obtained.

[0027] Moreover, formation of the stress relaxation layer which uses the grease mentioned above can be performed by dropping thru/or applying the grease of sufficient amount for a wrap for an organic EL device on an organic EL device, and forming a grease layer. Or after forming a grease layer as mentioned above, an accessory plate can be arranged on this grease layer, and it can carry out by spreading the aforementioned grease layer uniformly the weight of this accessory plate, or by applying a load to this accessory plate. Moreover, although it is also possible to form a stress relaxation layer by applying grease to mum order, without using an accessory plate, plastering thinly to mum order may damage an organic EL device, and it can be said to be difficult. And formation of the stress relaxation layer which uses the gel mentioned above can be performed like the case where a stress relaxation layer is formed using grease.

[0028] The thickness (thickness on an organic EL device) of the stress relaxation layer which can be formed as mentioned above is selectable suitably within the limits of 0.1 micrometers – 1cm in general. It is difficult for

thickness to acquire desired effectiveness is less than 0.1 micrometers. On the other hand, if thickness exceeds 1cm, since the advantage of an organic electroluminescence device called a thin shape is not only spoiled, but the increase of the volume by thermal expansion will become large according to increase of the volume of a stress relaxation layer, a possibility of destroying by the temperature change at the time of use etc. becomes strong. The thickness with a desirable stress relaxation layer is 0.01–1mm. In addition, thickness here means the thickness of the layer of the coating liquid which exists maintaining a liquid condition on the periphery of an organic EL device when forming using the coating liquid which the stress relaxation layer mentioned above.

[0029] Moreover, although this accessory plate may have the closure effectiveness and does not need to have it when using an accessory plate in forming a stress relaxation layer, it is more more desirable to have the closure effectiveness. It is desirable to use the tabular thing made as an accessory plate by resin, such as inorganic substances, such as glass, ceramics, and a metal, and fluororesin, acrylic resin, a polycarbonate, polyester, a polyamide, polystyrene, polypropylene, polyolefine system resin, etc. As mentioned above, when plane view of the magnitude of an accessory plate is carried out, it is desirable that it is sufficient magnitude for a wrap about an organic EL device. Moreover, as for the thickness of an accessory plate, it is desirable to be referred to in general as 0.1 micrometers – 3mm. The production cost of thickness is high, and a less than 0.1-micrometer thing does not have it from being hard to deal with it. [desirable] On the other hand, if thickness exceeds 3mm, the advantage of an organic electroluminescence device called a thin shape will be spoiled. The desirable thickness of an accessory plate is 0.01–1mm.

[0030] In the organic electroluminescence device of this invention, the stress relaxation layer which mentioned it above as covered the organic EL device formed on the substrate is prepared, and as the organic EL device covered with this stress relaxation layer is covered, the adhesives layer is formed on the aforementioned substrate. While it is for making the sealing agent mentioned later fix, this adhesives layer is for preventing that the stress relaxation layer mentioned above touches with the open air, and unless the configuration of the sealing agent which mentions later whether that side face prepares a special member is made special, it meets with the open air. Therefore, an adhesives layer has especially the desirable thing what has the closure effectiveness, i.e., water permeability, and oxygen permeability are [a thing] low. What consists of photoresist adhesives, thermosetting adhesive, anaerobic adhesive, etc. as such an adhesives layer is desirable, and BENEFIKKUSU VL (trade name of the photoresist adhesives Made from ADERU), acrylic one #4111 (trade name of the photoresist adhesives by Maruto Co., Ltd.), the ARON tightness UL (trade name of the anaerobic adhesive made from Toagosei Chemical industry), the AREMUKO bond 570 (trade name of the thermosetting adhesive Made from ODEKKU), etc. are mentioned as an example.

[0031] An adhesives layer trickles adhesives into the request part of a part in which the aforementioned stress relaxation layer is not prepared in the substrate front face of the side which prepared the stress relaxation layer top (on [when an accessory plate is used in forming a stress relaxation layer] this accessory plate) mentioned above, and this stress relaxation layer. The sealing agent besides mentioned later is arranged, and after spreading the aforementioned adhesives uniformly the weight of a sealing agent, or by applying a load to a sealing agent, it can form by stiffening the adhesives concerned. Moreover, after forming a non-hardened adhesives layer by approaches, such as the applying method, a spin coat method, and a dip method, and arranging the sealing agent later mentioned on this adhesives layer, it can form also by stiffening the adhesives concerned.

[0032] Final thickness of an adhesives layer (thickness on a stress relaxation layer.) When an accessory plate is used in forming a stress relaxation layer, as for the thickness on this accessory plate, it is desirable to be referred to in general as 0.1 micrometers – 1cm. When thickness carries a sealing agent in less than 0.1 micrometers, as a result of the sealing agent concerned coming floating with the surface tension of the adhesives in the condition of not hardening, air bubbles tend to go into the inside of the adhesives in the condition of not hardening, or the interface of this adhesives and sealing agent. On the other hand, if thickness exceeds 1cm, the advantage of an organic electroluminescence device called a thin shape will be spoiled. The thickness with a desirable adhesives layer is 0.001–1mm. Since the organic EL device is covered with the stress relaxation layer at the time of formation of an adhesives layer, there is no need of paying attention in the degree of hardness and contraction at the time of hardening of adhesives substantially. As the result, the width of face of the selection of adhesives from the case where there is no stress relaxation layer spreads.

[0033] Since the sealing agent which fixes by the adhesives layer mentioned above on a substrate (that in which the organic EL device is formed) is for moisture and oxygen to prevent invasion from the exterior to an organic EL device, it is desirable to use the high thing of the closure effectiveness, i.e., a permeable and oxygen penetrable small thing. As an example of the matter which can be used as a sealing agent, resin, such as inorganic substances, such as glass, ceramics, and a metal, and fluororesin, acrylic resin, a polycarbonate, polyester, a polyamide, polystyrene, polypropylene, polyolefine system resin, is mentioned.

[0034] Since the time and effort of processing is economical few, as for the configuration of a sealing agent, it is desirable that it is tabular. Moreover, although the thinner one of the thickness of a sealing agent is [from / in obtaining a thin organic electroluminescence device] desirable, since it will become weak to impacts, such as external force, if too thin, it is desirable to choose suitably within the limits of 0.1 micrometers – 1cm in general according to the quality of the material of a sealing agent. It is difficult to obtain that in which thickness has practically sufficient resistance to impacts, such as external force, in less than 0.1 micrometers. On the other hand, if thickness exceeds 1cm, it will become difficult to obtain a thin organic electroluminescence device. The more desirable thickness of a sealing agent is 0.01–1mm.

[0035] The organic electroluminescence device of this invention makes the organic EL device covered with the

stress relaxation layer mentioned above, the adhesives layer, and the aforementioned stress relaxation layer other than a sealing agent the requirements for an indispensable configuration. That an organic device should just be formed on a substrate, the lamination will not be limited, especially if it functions as an organic EL device.

[0036] As an example of the lamination of the organic EL device of the type which makes a substrate side the ejection side of light, the thing of following the (1) – (4) is mentioned for the order of a laminating on a substrate front face.

(1) An anode plate / luminous layer / cathode (2) anode plate / luminous layer / electron injection layer / cathode (3) anode plate / hole-injection layer / luminous layer / cathode (4) anode plate / hole-injection layer / luminous layer / electron injection layer / cathode [0037] Here, although a luminous layer is usually formed of one sort or two or more sorts of organic luminescent material, it may be formed with mixture with an organic luminescent material, a hole-injection ingredient, and/or an electron injection ingredient. Moreover, the protective layer for preventing invasion of the moisture to a component, as the component concerned is covered on the periphery of the component of lamination mentioned above may be prepared.

[0038] When making a substrate side into the ejection side of light, the tabular object which the aforementioned substrate consists of matter which gives high permeability (in general 80% or more) to luminescence (EL light) from an organic EL device at least, and specifically consists of clear glass, a transparent plastic, a quartz, etc., a sheet-like object, or a film-like object is used. In addition, it is also possible to make into the ejection side of light the sealing agent side mentioned above in the organic electroluminescence device of this invention. In this case, it is not necessary to make the order of a laminating on a substrate into above-mentioned sequence and above-mentioned reverse, to form an organic EL device, and to take into consideration the permeability of luminescence (EL light) from an organic EL device about a substrate ingredient. Instead, the matter which gives high permeability (in general 80% or more) to luminescence (EL light) from an organic EL device about the ingredient of a stress relaxation layer, the ingredient of an adhesives layer, and the ingredient of a sealing agent is used.

[0039] As an ingredient of an anode plate, cathode, a luminous layer, a hole-injection layer, an electron injection layer, and a protective layer, a well-known ingredient can be used conventionally, respectively. For example, the metal with a large (for example, 4eV or more) work function as an anode material, an alloy, electrical conductivity compounds, or such mixture are used preferably. As an example, metals, such as gold and nickel, dielectric transparent materials, such as CuI, ITO, SnO₂, and ZnO, etc. are mentioned. Especially, the point of productivity or a controllability to ITO is desirable. Although the thickness of an anode plate is based also on an ingredient, it is usually selectable suitably within the limits of 10nm – 1 micrometer.

[0040] Moreover, as a cathode material, a small (for example, 4eV or less) metal, an alloy, electrical conductivity compounds, or such mixture of a work function etc. are used preferably. As an example, rare earth metals, such as the alloy of a sodium and sodium-potassium alloy, magnesium, a lithium, magnesium, and silver or a mixed metal, aluminum, aluminum/AI0₂, an indium, and an ytterbium, etc. are mentioned. Although the thickness of an anode plate is based also on an ingredient, it is usually selectable suitably within the limits of 10nm – 1 micrometer. Also in any of an anode plate and cathode, below hundreds of ohms / ** of the sheet resistance are desirable. In addition, in case an anode material and a cathode material are chosen, the magnitude of the work function made into criteria is not limited to 4eV.

[0041] The impregnation function in which an electron can be poured in from cathode or an electron injection layer while the ingredient (organic luminescent material) of a luminous layer can pour in an electron hole from an anode plate or a hole-injection layer at the time of the luminous layer for organic EL devices, i.e., electric-field impression. What is necessary is just to be able to form the layer which has the transportation function to which the poured-in charge (an electron and an electron hole at least on the other hand) is moved by the force of electric field, the luminescence function to offer the place of the recombination of an electron and an electron hole and to tie this to luminescence, etc. As the example, the fluorescent brightener of systems, such as a benzothiazole system, a benzimidazole system, and a benzooxazole system, A metal chelation oxy-NOIDO compound, a styryl benzenoid compound, a JISUCHIRIRU pyrazine derivative, Polyphenyl system compound and 12-phtalo peri non, 1, 4-diphenyl-1,3-butadiene, 1, 1, 4, and 4-tetra-phenyl-1,3-butadiene, the North America Free Trade Agreement RUIMIDO derivative, The metal complex of a perylene derivative, an OKISA diazole derivative, an aldazine derivative, a PIRAJIRIN derivative, a cyclopentadiene derivative, a pyrrolo pyrrole derivative, a styryl amine derivative, a coumarin system compound, an aromatic dimethylidyne compound, and an eight-quinolinol derivative etc. is mentioned. Although especially the thickness of a luminous layer is not limited, it is usually suitably chosen within the limits of 5nm – 5 micrometers.

[0042] The ingredient (hole-injection ingredient) of a hole-injection layer should just have any of the impregnation nature of an electron hole, and electronic obstruction nature they are. As the example, a triazole derivative, an OKISA diazole derivative, An imidazole derivative, the poly aryl alkane derivative, a pyrazoline derivative, A pyrazolone derivative, a phenylenediamine derivative, an arylamine derivative, An amino permutation chalcone derivative, an oxazole derivative, a styryl anthracene derivative, full — me — non — a derivative, a hydrazone derivative, a stilbene derivative, and a silazane derivative — Conductive polymer oligomer, such as a polysilane system compound, an aniline system copolymer, and thiophene oligomer, a porphyrin compound, an aromatic series tertiary-amine compound, a styryl amine compound, an aromatic series JIMECHIRI DIN system compound, etc. are mentioned. Although especially the thickness of a hole-injection layer is not limited, either, it is usually suitably chosen within the limits of 5nm – 5 micrometers. A hole-injection layer may be 1 layer structure which consists of one sort of the ingredient mentioned above, or two sorts or more, and may be two or more layer structure which

consists of two or more layers of the same presentation or a different-species presentation. [0043] An electron injection layer that what is necessary is just to have the function to transmit the electron poured in from cathode to a luminous layer as an example of the ingredient (electron injection ingredient) nitration full — me — non — a derivative, an anthra quinodimethan derivative, and a diphenyl quinone derivative — Heterocycle tetracarboxylic acid anhydrides, such as a thiopyran dioxide derivative and naphthalene perylene, A carbodiimide, a deflection ORENIRIDEN methane derivative, an anthra quinodimethan derivative, A thing, a JISUCHIRIRU pyrazine derivative, etc. with which the metal complex of an anthrone derivative, an OKISA diazole derivative, and an eight-quinolinol derivative, a metal free phthalocyanine, metal phthalocyanines, or these ends are permuted by the alkyl group, the sulfone radical, etc. are mentioned. Although especially the thickness of an electron injection layer is not limited, either, it is usually suitably chosen within the limits of 5nm - 5 micrometers. An electron injection layer may be 1 layer structure which consists of one sort of the ingredient mentioned above, or two sorts or more, and may be two or more layer structure which consists of two or more layers of the same presentation or a different-species presentation.

[0044] And the copolymer which is made to carry out copolymerization of the monomer mixture containing tetrafluoroethylene and at least one sort of comonomers as an example of the ingredient of a protective layer, and is obtained, The fluorine-containing copolymer which has cyclic structure in a copolymerization principal chain, polyethylene, polypropylene, Polymethylmethacrylate, polyimide, polyurea, polytetrafluoroethylene, Polychlorotrifluoroethylene resin, poly dichlorodifluoroethene, The copolymer of chlorotrifluoroethylene and dichlorodifluoroethene, The absorptivity matter of 1% or more of water absorption, and the dampproof matter of 0.1% or less of water absorption, Metals, such as In, Sn, Pb, Au, Cu, Ag, aluminum, Ti, and nickel, MgO, SiO, SiO₂, aluminum 2O₃, GeO, NiO, CaO, BaO and Fe 2O₃, Y₂O₃, and TiO₂ etc. — a metallic oxide, MgF₂, LiF, AlF₃, and CaF₂ etc. — a metal fluoride etc. is mentioned.

[0045] Moreover, it is not limited especially about the formation approach of each class (an anode plate and cathode are included) which constitutes an organic EL device, either, as the formation approach of an anode plate, cathode, a luminous layer, a hole-injection layer, and an electron injection layer — a vacuum deposition method, a spin coat method, the cast method, the sputtering method, and LB — although law etc. is applicable, about a luminous layer, it is desirable to apply approaches other than the sputtering method (LB a vacuum deposition method, a spin coat method, the cast method, law, etc.). As for especially a luminous layer, it is desirable that it is the molecule deposition film. the thing of the thin film which deposition was carried out to the molecule deposition film from the ingredient compound of a gaseous-phase condition here, and was formed, and the film solidified and formed from the ingredient compound of a solution condition or a liquid phase condition — it is — usually — this molecule deposition film — LB — with the thin film (molecule built up film) formed of law, it is classifiable with the difference of condensation structure and higher order structure, and the functional difference resulting from it. In forming a luminous layer with a spin coat method etc., it prepares a coating solution by melting a binder and ingredient compounds, such as resin, to a solvent.

[0046] moreover — a protective layer — a vacuum deposition method, a spin coat method, the sputtering method, the cast method, and MBE (molecular beam epitaxy) — law, the ionized cluster beam method, the ion plating method, a plasma polymerization method (the high-frequency excitation ion plating method), a reactive-sputtering method, a plasma-CVD method, a laser CVD method, a heat CVD method, a gas source CVD method, etc. are applicable.

[0047] The formation approach of each class can be suitably changed according to the ingredient to be used. If a vacuum deposition method is used in formation of each class which constitutes an organic EL device, since an organic EL device can be formed only with this vacuum deposition method, it is advantageous when aiming at simplification of a facility, and compaction of the production time.

[0048] In addition, the configuration and magnitude of each class (a protective layer is included) which constitute an organic EL device are not necessarily the same. Moreover, when plane view of the component is carried out, all other layers are not surely settled on the electrode formed in right above [of a substrate]. On the other hand, the stress relaxation layer as used in the field of this invention is for controlling that the residual stress of an adhesives layer spreads to an organic EL device, and short-circuit arises in the organic EL device concerned. Therefore, "it is a wrap stress relaxation layer about an organic EL device" as used in the field of this invention means the stress relaxation layer which has covered at least the field with which cathode and an anode plate lap, when plane view of the organic EL device is carried out. Moreover, "it is the coating liquid (grease, gel) of sufficient amount for a wrap about an organic EL device" as used in this specification means "it is the coating liquid (grease, gel) of amount sufficient at least for a wrap about the field with which cathode and an anode plate lap when plane view of the organic EL device is carried out." And "the accessory plate which has sufficient magnitude for a wrap for an organic EL device when plane view is carried out" means "the accessory plate which has magnitude sufficient at least for a wrap for the field with which cathode and an anode plate lap when plane view is carried out, and plane view of the organic EL device is carried out."

[0049] Moreover, the number of the organic EL devices which constitute the organic electroluminescence device of this invention may be one, and may be plurality. When two or more, each component may have the same class seen from lamination or the luminescent color, and may differ. It is selectable suitably into how many the number of organic EL devices is made according to the application of the organic electroluminescence device made into the purpose etc. When two or more organic EL devices are formed on the substrate, a stress relaxation layer may be prepared for every organic EL device, one thing common to all organic EL devices may be prepared, and two or

more things common to the plurality [] the organic EL devices may be prepared. In for [] two or more organic EL devices formed on the substrate in a wrap sake for two or more stress relaxation layer [] the same is said of the case where an accessory plate is used, and one thing common to [] may use an accessory plate for every stress relaxation layer, and [] all stress relaxation layers which it is going to prepare may be prepared, and two or more things common to the plurality of all the stress relaxation layers may be prepared. About an adhesives layer and a sealing agent, even when two or more organic EL devices are formed on the substrate, it is desirable practically to prepare one thing common to all organic EL devices, respectively.

[0050] the organic EL device which mentioned above the organic electroluminescence device of this invention, and this organic EL device — a wrap — the above-mentioned stress relaxation layer which made it like and was formed on the substrate (that in which the organic EL device is formed), and this stress relaxation layer — a wrap — it has the above-mentioned adhesives layer which made it like and was formed on the aforementioned substrate, and the above-mentioned sealing agent which fixed on the aforementioned substrate by this adhesives layer. In this organic electroluminescence device, since the organic EL device is covered with the stress relaxation layer, it is controlled that the residual stress of an adhesives layer spreads to an organic EL device. Since it is prevented that the crush of the edge of an organic EL device etc. is carried out by the residual stress spread from the adhesives layer, and it short-circuits with it as the result, the luminescence function of an organic EL device continues at a long period of time, and it is hard to lose it.

[0051] Moreover, although the force works in the parallel direction with the front face of an organic EL device as usual in the organic electroluminescence device of this invention at the time of formation of an adhesives layer, it is controlled from the organic EL device being covered with the stress relaxation layer that the aforementioned force spreads to an organic EL device. Since it is prevented by the organic EL device according to the aforementioned force as the result that electrode exfoliation occurs, it cannot break out easily that the luminescence function of an organic EL device loses after the manufacture process of an organic electroluminescence device or manufacture. Furthermore, since the organic electroluminescence device of this invention closes an organic EL device by the adhesion type closure approach, its closure effectiveness of an organic EL device is high, and its thickness of the device itself is thin, and it is comparatively easy to mass-produce.

[0052] The organic electroluminescence device of this invention which has such a description can be used as the surface light source, a character indicating equipment, the equipment for electric spectacles, the indicator for mount, the light source for electric discharge of a copying machine, the light source for printers, light modulation equipment, etc.

[0053] Next, the closure approach of the organic EL device of this invention is explained. This approach is characterized by preparing an adhesives layer in it, as this organic EL device is covered at the tooth back of an organic EL device prepared on the substrate, and forming said organic EL device for a wrap stress relaxation layer between said organic EL devices and said adhesives layers in closing said organic EL device by fixing a sealing agent on said substrate by this adhesives layer, as mentioned above.

[0054] Preparing in order to control that the residual stress of an adhesives layer spreads the aforementioned stress relaxation layer to an organic EL device, the ingredient and the formation approach are as having explained in the organic electroluminescence device of this invention mentioned above. Moreover, it is as having also explained the detail of the closure which fixes the organic EL device covered with this stress relaxation layer, and the organic EL device covered with the aforementioned stress relaxation layer by the adhesives layer prepared as covers, and the aforementioned adhesives layer on a substrate (that in which the organic EL device is formed) in the organic electroluminescence device of this invention mentioned above, respectively.

[0055] In the organic EL device closed by this approach, since the organic EL device concerned is covered with the stress relaxation layer, it is controlled that the residual stress of an adhesives layer spreads to an organic EL device. Since it is prevented that the crush of the edge of an organic EL device etc. is carried out by the residual stress spread from the adhesives layer, and it short-circuits with it as the result, the organic EL device (that by which the closure was carried out (= organic electroluminescence device)) which a luminescence function continues at a long period of time, and cannot lose easily is obtained.

[0056] Moreover, although the force works in the direction parallel to the front face of an organic EL device at the time of formation of an adhesives layer, it is controlled from the organic EL device being covered with the stress relaxation layer that the aforementioned force spreads to an organic EL device. Since it is prevented by the organic EL device according to the aforementioned force as the result that electrode exfoliation occurs, it cannot break out easily that the luminescence function of an organic EL device loses after the process of the closure or the closure. Furthermore, since the approach of this invention is one of the adhesion type closure approaches, it is comparatively easy for the closure effectiveness to be high and to mass-produce an organic EL device (that by which the closure was carried out (= organic electroluminescence device)) with thin thickness.

[Translation done.]

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EXAMPLE

[Example] Hereafter, the example of this invention is explained.

After preparing what formed the ITO film of 100nm of thickness to band-like at the longitudinal direction of the glass substrate concerned (this is hereafter called transparency support substrate) on production place ** of an example 1 (1) organic EL device, and the glass substrate of 25mmx75mmx1.1mm magnitude and cleaning this transparency support substrate ultrasonically for 30 minutes by isopropyl alcohol, pure water washed for 30 minutes and, finally it cleaned ultrasonically for 30 minutes by isopropyl alcohol again. The transparency support substrate after washing is fixed to the substrate electrode holder of a commercial vacuum evaporation system (product made from Japanese Vacuum technology). On the resistance heating boat made from molybdenum, N, N'-diphenyl-N, N'-screw-(3-methylphenyl)-[1 and 1'-biphenyl]-4, and 4'-diamine It put in (it having been hereafter called TPD), 200mg (henceforth Alq3) of tris (eight quinolinol) aluminum was put into another resistance heating boat made from molybdenum, and the inside of a vacuum chamber was decompressed up to 1x10 to 4 Pa. [200mg]

[0058] Next, heated the aforementioned resistance heating boat into which TPD was put to 215-220 degrees C, TPD was made to deposit on the ITO film in the evaporation rate of 0.1-0.3nm/second, and the hole-injection layer of 60nm of thickness was formed. The substrate temperature at this time was a room temperature. Subsequently, the luminous layer was successively formed to membrane formation of a hole-injection layer, without taking out the transparency support substrate with which the hole-injection layer was formed from a vacuum chamber. Membrane formation of a luminous layer is Alq3. The put-in aforementioned resistance heating boat is heated to 275 degrees C, and it is Alq3. It is made to deposit on a hole-injection layer in the evaporation rate of 0.1-0.2nm/second, and is Alq3 of 60nm of thickness. It carried out by forming a layer. The substrate temperature at this time was also a room temperature. Next, magnesium 1g was put into the resistance heating boat made from molybdenum, 500mg of silver was put into another resistance heating boat made from molybdenum, and the inside of a vacuum chamber was decompressed up to 2x10 to 4 Pa. And while heating the aforementioned resistance heating boat into which magnesium was put at about 500 degrees C and evaporating magnesium in the evaporation rate of about 1.7-2.8nm/second, the aforementioned resistance heating boat into which silver was put was heated at about 800 degrees C, silver was evaporated in the evaporation rate of 0.03-0.08nm/second, and the cathode of 150nm of thickness which consists of a mixed metal of magnesium and silver was established in a total of on [three] a luminous layer. The magnitude on plane view of each cathode is 3mmx15mm, and these are prepared in the sense which intersects perpendicularly with the ITO film at fixed spacing.

[0059] Then, the glass substrate prepared to the aforementioned cathode was cut to three, and a total of three organic EL devices whose lamination on a glass substrate is an anode plate (ITO film) / hole-injection layer / luminous layer / cathode (Mg-Ag layer) were obtained. The magnitude of the luminescence side (field where cathode and an anode plate lap in when plane view is carried out, and the hole-injection layer and the luminous layer exist among these electrodes) of these organic EL devices is 3mmx5mm. Moreover, the initial brightness of these organic EL devices is electrical-potential-difference 6.5V and current density 3 mA/cm². They are 100 cd/m² under conditions. Reaching, the power conversion effectiveness at this time was 1.6 lm/W.

[0060] (2) the formation list of a stress relaxation layer and an adhesives layer — as the ingredient of arrangement place ** of a sealing agent, and a stress relaxation layer — fluororesin coating liquid (SAITOPPU CTX-105A by Asahi Glass Co., Ltd.) — preparing — moreover — as an accessory plate — about 10 — the cover glass (henceforth cover glass I) of mmx10mmx0.15mm magnitude was prepared. Next, it was dropped on the organic EL device which produced the 5micro of the aforementioned fluororesin coating liquid I above (1), and the aforementioned cover glass I was carried on this. The thin film (0.05mm in thickness) of said coating liquid was uniformly made by said coating liquid with the viscous lowness of fluororesin coating liquid, and the weight of cover glass I between breadth, an organic EL device, and cover glass I. At this time, some coating liquid was protruded into the outside of the field covered by cover glass I a little. By leaving it about 10 minutes in this condition, the solid-state layer (coat) was made to form in the outside surface of the above-mentioned thin film which consists of fluororesin coating liquid, i.e., the front face of a glass substrate and the front face of an parallel direction, and the stress relaxation layer made into the purpose was obtained once. At this time, the fluororesin coating liquid of the part which is in contact with the organic EL device was maintaining the liquid condition, without solidifying.

[0061] Next, 0.1ml (BENEFIKKUSU VL Made from ADERU) of photoresist adhesives was dropped on the above-mentioned cover glass I as adhesives, and the cover glass (magnitude is called cover glass II below about 20mmx20mmx0.15mm;) as a sealing agent was carried on it. Said photoresist adhesives covered completely breadth, the above-mentioned stress relaxation layer, and cover glass I uniformly with the viscous lowness of photoresist

adhesives, and the weight of cover glass I. Following on installation of cover glass II, the light of a halogen lamp was irradiated from on the aforementioned cover glass II at photoresist adhesives, and the photoresist adhesives concerned were stiffened.

[0062] The adhesives layer with a thickness of 0.1mm (thickness on cover glass I) which consists of photoresist adhesives by this as covers the above-mentioned stress relaxation layer was formed, and the cover glass II as a sealing agent fixed by the aforementioned adhesives layer to coincidence on the glass substrate (that in which the organic EL device is formed). Furthermore, since the vaporization of the solvent of the fluororesin coating liquid in the above-mentioned stress relaxation layer was inhibited after forming an adhesives layer, finally the stress relaxation layer to which it continues maintaining the fluororesin coating liquid which exists in the periphery of an organic EL device, and it targets a liquid condition as the result was formed.

[0063] By having carried out to formation of an adhesives layer, and arrangement of a sealing agent as mentioned above, the organic electroluminescence device made into the purpose was obtained. Hereafter, the organic electroluminescence device was produced similarly and a total of 45 organic electroluminescence devices were obtained. The schematic diagram of the cross section of the organic electroluminescence device produced by this example 1 is shown in drawing 1 R>1, and a plan is shown in drawing 2 .

[0064] The organic EL device 3 with which this organic electroluminescence device 1 was formed in one side of a glass substrate 2 as shown in drawing 1 . The stress relaxation layer 4 formed on the glass substrate 2 as covered this organic EL device 3. The accessory plate 5 which consists of cover glass I used in order to form this stress relaxation layer 4. It has the adhesives layer 6 which consists of photoresist adhesives formed on the glass substrate 2 as covered this accessory plate 5 and said stress relaxation layer 4, and the sealing agent 7 which consists of cover glass II which fixed on the glass substrate 2 by this adhesives layer 6. The stress relaxation layer 4 is made from fluororesin coating liquid, in this stress relaxation layer 4, the front face of a glass substrate 2 and the front face of a parallel direction are solidified, when the solvent evaporated at the time of that formation, solid-state layer 4a is formed, and fluororesin coating liquid 4b of the part which is in contact with the organic EL device 3 is maintaining the liquid condition, without solidifying.

[0065] Moreover, an organic EL device 3 is Alq3 as the ITO film as an anode plate, the TPD layer as a hole-injection layer, and a luminous layer on a glass substrate 2. The laminating of a layer and the Mg—Ag mixolumnion as cathode is carried out one by one. It is sign 3a about the ITO film among these, and sign 3b shows a Mg—Ag mixolumnion in drawing 1 and drawing 2 , respectively. As shown in drawing 1 and drawing 2 , ITO film 3a is formed in glass substrate 2 front face band-like, and Mg—Ag mixolumnion 3b is prepared in the sense which intersects perpendicularly with ITO film 3a. And the part with which ITO film 3a and Mg—In mixolumnion 3b have lapped on plane view is equivalent to the luminescence side of an organic EL device 3.

[0066] an example 2 — the organic EL device was first produced on the glass substrate on an example 1 and these conditions. next, the this top after applying these fluorine system grease about 0.01ml on the above-mentioned organic EL device, using fluorine system grease (PTFE grease by NICHIAS CORP.) as an ingredient of a stress relaxation layer — as an accessory plate — about 10 — the cover glass I of mmx10mmx0.15mm magnitude was carried. Subsequently, by pressing the aforementioned cover glass lightly from a top, the aforementioned fluorine system grease was spread uniformly and the thin film (0.1mm in thickness) of fluorine system grease was formed between an organic EL device and cover glass I. Thereby, the stress relaxation layer was obtained. Then, formation of the adhesives layer (0.06mm in thickness) which consists of photoresist adhesives on an example 1 and these conditions, and fixing of the sealing agent (cover glass II) by this adhesives layer were performed, and the organic electroluminescence device made into the purpose was obtained. Hereafter, the organic electroluminescence device was produced similarly and a total of 55 organic electroluminescence devices were obtained.

[0067] an example 3 — the organic EL device was first produced on the glass substrate on an example 1 and these conditions. Next, the aluminum layer of 0.2 micrometers of thickness was formed with the vacuum deposition method as a protective layer on the cathode (Mg—Ag layer) which constitutes the aforementioned organic EL device. Then, formation of the stress relaxation layer (0.05mm in thickness) using an accessory plate (cover glass I), formation of the adhesives layer (0.1mm in thickness) which consists of photoresist adhesives, and fixing of the sealing agent (cover glass II) by this adhesives layer were performed on an example 1 and these conditions, and the organic electroluminescence device made into the purpose was obtained. Hereafter, the organic electroluminescence device was produced similarly and a total of 45 organic electroluminescence devices were obtained.

[0068] A total of 45 organic electroluminescence devices were produced like the example 1 except having not formed an example of comparison 1 stress-relaxation layer. An accessory plate was not used at this time, either.

[0069] A total of 45 organic electroluminescence devices were produced like the example 1 except both having not fixed the sealing agent by formation of an example of comparison 2 adhesives layer, and the adhesives layer.

[0070] the example 3 of a comparison — after washing this transparence support substrate on an example 1 and these conditions first, using that by which the 10mmx75mmx100nm ITO film was formed by the edge of both directions of a short hand of one side of the glass substrate of 25mmx75mmx1.1mm magnitude, respectively as a transparence support substrate, the substrate electrode holder of the vacuum evaporation system equipped with the mask automatic switchboard style was equipped. Next, the mask was covered over one side of the above-mentioned ITO film, and the hole-injection layer and the luminous layer were formed on an example 1 and these conditions by this condition, respectively. Subsequently, after removing the above-mentioned mask using the mask automatic switchboard style attached to vacuum evaporation equipment, width of face of 5mm was covered at the

radial-border section of both longitudinal directions of one ITO film (ITO film of the direction over which the aforementioned mask was covered), respectively as it is also at said device, and the mask was covered.

[0071] Next, heat the resistance heating boat made from molybdenum into which magnesium 1g was put beforehand, and the resistance heating boat made from molybdenum into which 500mg of silver was put beforehand, respectively, made magnesium vapor-deposit with the evaporation rate of about 1.5nm/second, coincidence was made to vapor-deposit silver with the evaporation rate of about 0.1nm/second, and the cathode of 150nm of thickness which consists of a mixed metal of magnesium and silver was formed on the luminous layer. By having formed membranes to cathode, the organic EL device was formed on the above-mentioned glass substrate.

[0072] Then, the protective layer was formed in the way of the following [the bottom of a series of vacuum environments which continue from membrane formation of a hole-injection layer, a luminous layer, and cathode], using the above-mentioned vacuum evaporation system as it is. First, the crucible made from an alumina which held 1.5g (Du Pont Teflon AF) of amorphism copolymer powder of tetrafluoroethylene, and perfluoro -2, the 2-dimethyl -1 and 3-JIOKI SOL as an evaporation source is beforehand put into the basket made from a tungsten, and the mesh made from stainless steel of 12 micrometerphi was put above this basket made from a tungsten (on the crucible made from an alumina). Subsequently, after decompressing the inside of a vacuum chamber up to 1×10^{-4} Pa, energization heating was carried out at the aforementioned basket made from a tungsten, the aforementioned evaporation source was heated at 455 degrees C, and the protective layer (Teflon AF thin film) of 800nm of thickness was formed with the evaporation rate of 0.5nm/second on the above-mentioned cathode which constitutes an organic EL device.

[0073] Then, the organic EL device which formed membranes to the protective layer was taken out from the vacuum chamber, the casing type closure was performed in the following ways about this organic EL device, and the organic electroluminescence device was obtained. First, the hole-injection layer which covers width of face of 5mm, respectively, and is prepared on this ITO film about the radial-border section of both longitudinal directions of the ITO film of the direction over which a mask was not covered in membrane formation of a hole-injection layer, a luminous layer, cathode, and a protective layer were excised. Moreover, also about the edge of both directions of a short hand of a glass substrate, width of face of 5mm was covered, respectively, and a hole-injection layer, a luminous layer, cathode, and a protective layer were excised so that the thickness might become the sum of the thickness of a glass substrate, and the thickness of the ITO film substantially.

[0074] Subsequently, the glass plate (outside **: call it 20mmx75mmx3mm and following shield glass) which has a 18mmx73mmx2mm crevice and a through tube (henceforth an inlet) with a diameter of 2mm prepared in the bottom of this crevice is prepared, and this shield glass and the organic EL device which formed membranes to the protective layer were stuck with epoxy system adhesives (Cemedine super by Cemedine Co., Ltd. 5). After epoxy system adhesives mixed base resin and a curing agent and stirred them 20 times by the spatula, they were applied to the rectangle of about 20x75mm by width of face of 1mm at the edge of the organic EL device which formed membranes to the above-mentioned protective layer. Moreover, into the crevice of shield glass, cathode and a protective layer stuck it, as shield glass and the organic EL device which formed membranes to the above-mentioned protective layer were settled. After lamination and in atmospheric air, it was left for 10 hours and epoxy system adhesives were stiffened.

[0075] Subsequently, the silicone oil (it is called TSK451 by Toshiba Corp. and the liquid for the following closures) which did 8 volume % distribution of the silica gel for moisture absorption (particle size of 50 micrometers) was poured in from the inlet established in shield glass, and the inside of the space formed by the crevice of shield glass and the organic EL device which formed membranes to the above-mentioned protective layer was filled with the liquid for the closures. Then, the glass lid blocked the inlet and the organic electroluminescence device with which the casing type closure was given was obtained. In addition, the glass lid was pasted up on shield glass with the epoxy system adhesives mentioned above. Hereafter, the organic electroluminescence device was produced similarly and a total of 45 organic electroluminescence devices were obtained.

[0076] About the organic electroluminescence device produced, respectively in the evaluation example 1 of the yield - the example 3 and the example 1 of a comparison - the example 3 of a comparison, immediately after device production to initial brightness is 100 cd/m². The continuation drive of the organic EL device was carried out by the becoming direct-current constant current, and the number of the organic EL devices which lost the luminescence function more short at each time one week, one month (for 31 days), and 1000 hours after [immediately after continuation drive initiation and] was counted. In addition, measurement of brightness was performed in atmospheric air using the luminance meter (CS-100 by Minolta Co., Ltd.). This result is shown in Table 1.

[0077]

[Table 1]

表 1

	評価に付した デバイスの数	発光機能を喪失した素子の数			
		駆動開始直後	1週間後	1箇月後	1000時間後
実施例 1	45	1	1	1	1
実施例 2	45	3	3	3	3
実施例 3	45	4	4	4	4
比較例 1	45	43	45	45	45
比較例 2	45	2	8	31	45
比較例 3	45	1	2	2	3

[0078] As shown in Table 1, there are few organic EL devices which lost the luminescence function more short in the organic electroluminescence device produced in the example 1 – the example 3, respectively. This shows that the reliable organic electroluminescence device was produced under the high yield. On the other hand, in the example 1 of a comparison which did not prepare a stress relaxation layer, almost all organic EL devices had lost the luminescence function more short at the time immediately after continuation drive initiation, and all the organic EL devices had lost the luminescence function more short after one week. This shows that a stress relaxation layer is useful, when an organic EL device prevents losing the luminescence function therefore short.

[0079] Moreover, in the organic electroluminescence device produced in the example 2 of a comparison, although there were few organic EL devices which therefore lost the luminescence function short at the time immediately after continuation drive initiation, 1000 hours after, all the components had lost the luminescence function more short. And in the organic electroluminescence device produced in the example 3 of a comparison, it is at all the investigation time and is [whether the number of the organic EL devices which therefore lost the luminescence function short is equivalent to an example 1 – an example 3, and] less than [it]. Therefore, it can say that the organic electroluminescence device with the high dependability seen from a viewpoint whether an organic EL device therefore loses that luminescence function short was produced under the high yield, and the organic electroluminescence device of the example 3 of a comparison is excellent in the example 3 of a comparison with this point. However, if the closure effectiveness mentioned later, the difficulty of manufacture, the thickness of an organic electroluminescence device, etc. are taken into consideration, it can be said that the direction of the organic electroluminescence device of an example 1 – an example 3 is excellent.

[0080] The round formal field called the dark spot looked at by the evaluation organic EL device of the closure effectiveness non-emitted light is expanded to the organic EL device, when moisture and oxygen invade. Therefore, it is suitable as an index for evaluating the closure effectiveness to investigate aging of the magnitude about the dark spot of arbitration. Immediately after the continuation drive initiation for the evaluation of the yield mentioned above from such a viewpoint, A photograph of each organic electroluminescence device is taken at each [of one day, one week, one month (for 31 days), and 1000 hours after] time. One device was chosen as arbitration from the organic electroluminescence devices which had not lost the luminescence function at the time 1000 hours after, and it asked for each diameter in a time from the aforementioned photograph paying attention to the dark spot of the arbitration in the organic EL device which constitutes the organic electroluminescence device. This result is shown in Table 2. In addition; since all the organic EL devices had lost the luminescence function more short about the organic electroluminescence device produced in the example 1 of a comparison at the time one week after, one device was chosen as arbitration from the organic electroluminescence devices which had not lost the luminescence function at the time one day after. Moreover, since all the organic EL devices had lost the luminescence function more short about the organic electroluminescence device produced in the example 2 of a comparison at the time 1000 hours after, one device was chosen as arbitration from the organic electroluminescence devices which had not lost the luminescence function at the time one month after.

[0081]

[Table 2]

表 2

	駆動開始直後	1 日 後	1 週 間 後	1 箇 月 後	1000 時間後
実施例 1	1 4	1 7 (21%)	1 8 (29%)	2 1 (50%)	2 3 (64%)
実施例 2	1 1	1 5 (36%)	1 9 (73%)	2 3 (109%)	2 5 (127%)
実施例 3	6	8 (33%)	9 (50%)	1 2 (100%)	1 3 (117%)
比較例 1	1 3	1 6 (23%)	—	—	—
比較例 2	1 7	3 2 (88%)	8 0 (371%)	450 (2550%)	—
比較例 3	1 2	1 5 (25%)	2 5 (108%)	3 0 (150%)	3 3 (175%)

*表内の数字の単位は μm

*括弧内の数値は駆動開始直後のダークスポットの直径に対する直径の増大分を百分率で表したものである。

*実線を付した空欄は、全ての素子が発光機能を喪失したために測定できなかったことを示す。

[0082] As shown in Table 2, also after carrying out a continuation drive for 1000 hours, with the organic electroluminescence device produced in the example 1 – the example 3, respectively, the dark spot is not expanded so much. This shows that the high closure effectiveness is acquired to an organic EL device in each organic electroluminescence device of an example 1 – an example 3. O Sufficient data for the organic electroluminescence device of the example 1 of a comparison to estimate the closure effectiveness on the other hand, since the organic EL device which loses a luminescence function more short succeeded one another were not able to be obtained. Moreover, it turns out that growth of a dark spot is very quick and the organic electroluminescence device of the example 2 of a comparison of the closure to an organic EL device is inadequate. And in the organic electroluminescence device of the example 3 of a comparison, growth of a dark spot is a little quick, and the closure effectiveness over an organic EL device understands that nearby [of an example 1 – an example 3] is low.

[Translation done.]

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]**[0085]**

[Drawing 1] It is the schematic diagram showing the cross section of the organic electroluminescence device produced in the example 1.

[0086]

[Drawing 2] It is the schematic diagram showing the top face of the organic electroluminescence device produced in the example 1.

[0087]**[Description of Notations]**

1 Organic Electroluminescence Device

2 Glass Substrate

3 Organic EL Device

4 Stress Relaxation Layer

5 Accessory Plate

6 Adhesives Layer

7 Sealing Agent

[Translation done.]

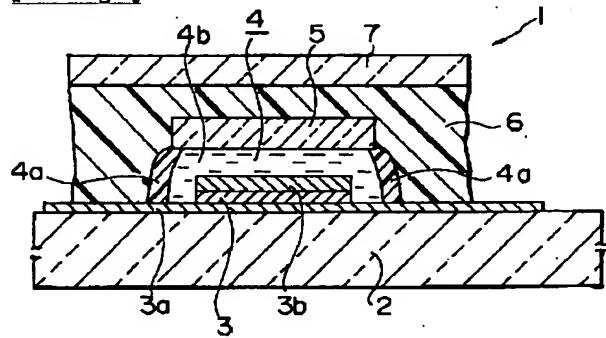
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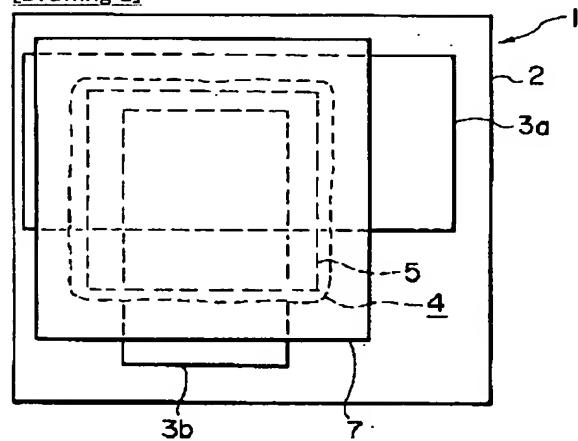
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DRAWINGS

[Drawing 1]



[Drawing 2]



[Translation done.]
